Temporal and spatial distribution of plutonium released from Nagasaki atomic bomb

（長崎原爆により放出されたプルトニウムの時間的及び空間的分布）

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Abstract

The Nagasaki atomic bomb was detonated on August 9, 1945. Following the detonation, plutonium and fission products were dispersed over the area of Nagasaki. The plutonium isotopes, ^{239}Pu and ^{240}Pu, have long half-lives and remain in the environment long term. Study of the plutonium in the environment provides information for understanding of behavior and migration of plutonium over a long period of time, because the source of the release was evident and Nagasaki is one of the oldest cities where plutonium was deposited.

Since 1945, atmospheric nuclear tests have been carried out worldwide, which resulted in additional plutonium inventory as global fallout. Thus plutonium deposited in Nagasaki area became a mixture of the plutonium from the atomic bomb and that from the global fallout. Discrimination of the plutonium due to atomic explosion in Nagasaki from global fallout plutonium is required in order to understand the behavior of the plutonium from the atomic bomb. Since the isotopic composition of plutonium depends on its origin, $^{240}Pu/^{239}Pu$ ratio is an effective tool for the identification of the source of plutonium.

In this paper, the $^{240}Pu/^{239}Pu$ ratios in sediments and surface soils were examined with a double-focusing inductively coupled plasma mass spectrometer. The data are applied to identify the temporal and spatial distribution of the plutonium from the atomic bomb.

Firstly, sediment cores were collected at Nishiyama reservoir in Nagasaki city, which experienced the 'black rain' formed from the detonation of the atomic bomb, and the depth profiles of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ and ^{137}Cs concentrations in the cores were determined to obtain depositional record of plutonium and ^{137}Cs released from the atomic bomb for 60 years. Identified were sediments containing deposition from the atomic bomb accumulated in 1945 and that from the nuclear tests in 1963, which resulted in chronological markers in the core. The profiles also revealed that the plutonium and ^{137}Cs from the atomic bomb were deposited abruptly after the detonation and, even at present, they were continuously being deposited together with those from the nuclear tests. By using two

chronological markers, other events such as a flood and a fire were observed in the core.

Secondly, the $^{240}Pu/^{239}Pu$ ratio and the $^{239+240}Pu$ and ^{137}Cs concentrations were measured in surface soils collected within a 10 km radius of the hypocenter of the atomic bomb. The $^{240}Pu/^{239}Pu$ ratio in this eastern area was lower than global fallout value. This finding clearly indicated that the plutonium was derived from the atomic bomb exploded over Nagasaki and dispersed still further away.

In order to determine the entire distribution of plutonium from the atomic bomb, the study area was expanded toward the eastern area of Nagasaki city. The $^{239+240}Pu$ concentrations in soils, except for Nishiyama area, were close to the average values for soils collected in Japan. In soils collected at the Nishiyama area and at the eastern area of Nagasaki prefecture, and even at part of northern area in Kumamoto prefecture, the $^{240}Pu/^{239}Pu$ ratios were lower than the global fallout value. This disclosed that the plutonium from the atomic bomb was deposited between about 2 km and about 100 km east from the hypocenter and about 30 km north and south wide. This distribution suggested that the deposition of the plutonium might be affected by wind, precipitation and geographical features.

It was also found that the isotopic ratio is more effective in identifying the origin of plutonium, than the methods based on $^{239+240}Pu/^{137}Cs$ activity ratio and $^{239+240}Pu$ and ^{137}Cs concentrations.

In conclusion, the results of temporal distribution connected with those of the spatial distribution permitted us to understand behavior of the plutonium released from the atomic bomb from its release to sedimentation.

Contents

List of Table

List of Photo

List of Figure

1. Introduction

The plutonium atomic bomb (called 'Fatman') exploded above Nagasaki city at 11:02 a.m. on August 9, 1945, 3 days after the detonation at Hiroshima. The explosion occurred at 503 m above the ground and caused approximately 150 thousand casualties and the physical damages in the city (Fig. 1-1). About 1.2 kg of the plutonium fissioned (Hansen and Paxton, 1969) and the remainder (13.8 kg) became a constituent of the fallout together with fission products, as well as activation products from the bomb fragments and other substances in the air or on the earth. Kimura et al. (1953) reported that soils collected after 1 month of the detonation at Nishiyama area (about 3 km east from the hypocenter), which experienced the 'black rain' formed from the detonation of the Nagasaki atomic bomb, had ^{89}Sr ($T_{1/2}$: 52.7 d), ^{140}Ba (12.8 d), ^{144}Ce (284 d)–^{144}Pr (17.3 min) and ^{95}Zr (65.5 d). In subsequent studies, found in soils collected at this area were plutonium isotopes (^{239}Pu: 2.41×10^4 y, ^{240}Pu: 6.564×10^3 y) and fission products such as ^{137}Cs (30.07 y), ^{90}Sr (28.78 y), and activation products such as ^{60}Co (5.281 y), ^{152}Eu (13.54 y) (e.g., $^{239+240}Pu$, ^{90}Sr: Sakanoue and Tsuji, 1971; ^{137}Cs: Okajima et al., 1978; ^{60}Co: Shizuma et al., 2002a; ^{152}Eu: Nakanishi et al., 1998; Shizuma et al., 2002b). Although fission products and activation products with relatively short half-lives decay out, plutonium isotopes have long half-lives and are kept in the environment over the long term. The plutonium released from the atomic bomb is interesting in research on environmental radioactivity, because evident information of the plutonium release provide opportunities to simplify understanding of environmental behavior of plutonium and the Nagasaki area is one of the oldest areas that were contaminated with the artificial plutonium in the world.

The released plutonium goes through several routes and migrates in the environment. It is said that most of plutonium moves to sediment in hydrosphere and then accumulates in the sediment. Thus, a reservoir plays the part of a final sink for the fallout from the atomic bomb in the basin. Nishiyama reservoir located at the center of the Nishiyama area is attractive to researchers. The sediments in the reservoir enable us to understand chronological trend of the deposit for about 100 years, because the reservoir was built in 1904 and there is, at least, no

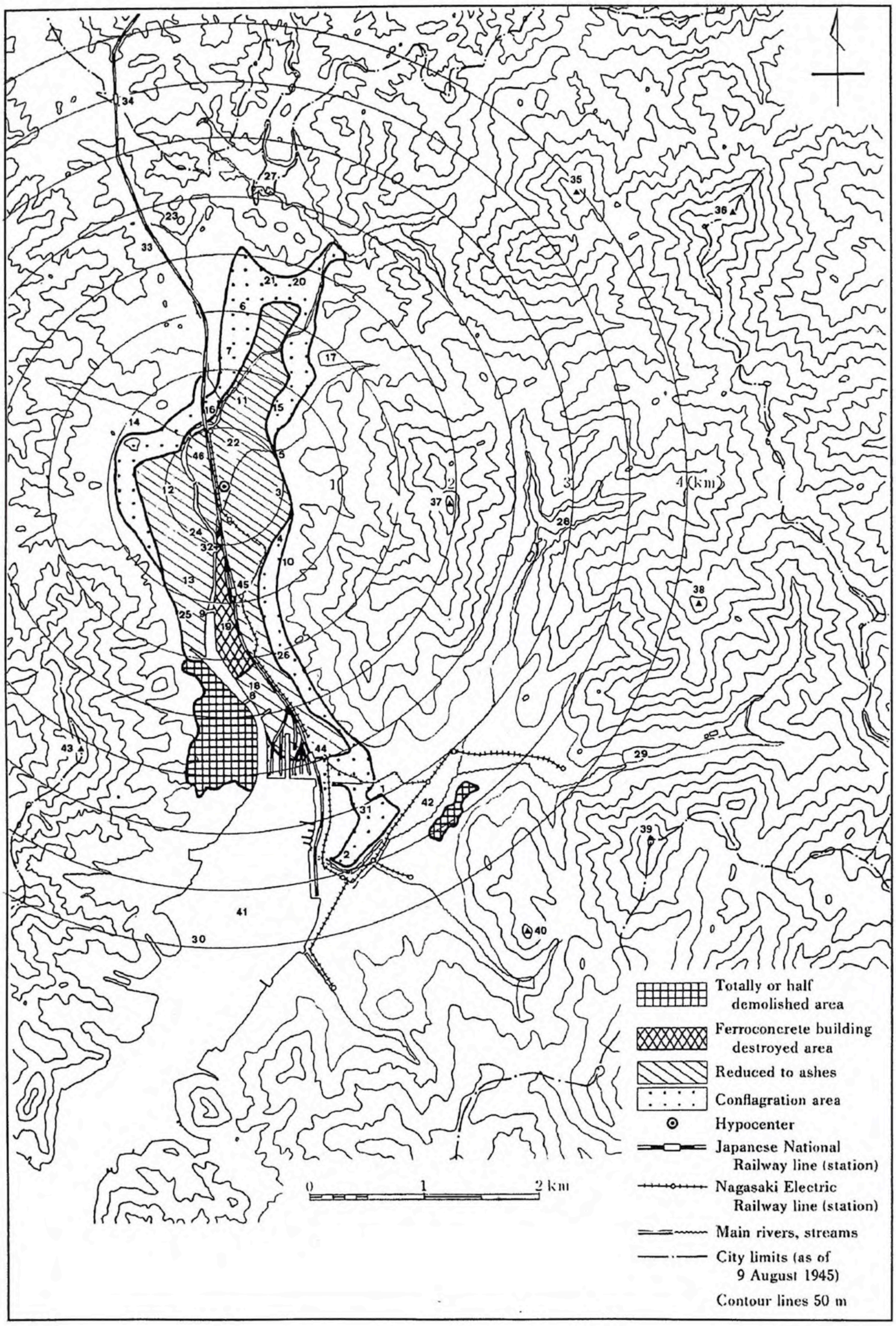

Fig. 1-1 State of damage to buildings in Nagasaki after detonation of Nagasaki atomic bomb. (The Committee for the Compilation of Materials on Damage Caused by the Atomic Bombs in Hiroshima and Nagasaki, 1981)

1. Nagasaki City Office
2. Nagasaki Prefectural Office
3. Nagasaki Medical University
4. Nagasaki Medical University Hospital
5. Urakami Cathedral
6. Mitsubishi Heavy Industries (MHI) Nagasaki Ordnance Factory, Ohashi Plant
7. MHI Nagasaki Shipyard, Ohashi Parts Plant
8. Inasa Bridge
9. Yanagawa Bridge
10. Sanno Shrine
11. Yamazato Primary School
12. Shiroyama Primary School
13. Keiho Middle School
14. Nagasaki Commercial School
15. Nagasaki Technical School
16. Ohashi Bridge
17. Urakami First Hospital
18. MHI Nagasaki Shipyard, Saiwaimachi Plant
19. Mitsubishi Steel Manufacturing Co. First Nagasaki Plant
20. Nishi-Urakami Primary School
21. Nagasaki Normal School
22. Urakami Prison
23. MHI Nagasaki Ordnance Factory, Sumiyoshi Tunneling Works
24. Chinzei Middle School
25. Fuchi Primary School
26. Zenza Primary School
27. Urakami Reservoir
28. Nishiyama Reservoir
29. Hongochi Reservoir
30. MHI Nagasaki Shipyard
31. Shinkozen Primary School
32. Mitsubishi Electric Corporation Nagasaki Foundry
33. Nagasaki Main Line
34. Michinoo Station
35. Mt. Tenjiku
36. Mt. Hoba
37. Mt. Konpira
38. Mt. Hoka
39. Mt. Higo
40. Mt. Atago
41. Nagasaki Port
42. Nakashima River
43. Mt. Inasa
44. Nagasaki Station
45. Urakami Station
46. Urakami River

record of dredging since its construction.

Sakanoue (1987) and Mahara et al. (1988) reported depth profiles of $^{239+240}$Pu and ^{137}Cs concentrations in sediments of the Nishiyama reservoir. Sakanoue suggested that the peak of $^{239+240}$Pu concentration was related to deposition of plutonium from the atomic bomb and Mahara et al. reported that high levels of $^{239+240}$Pu and ^{137}Cs in sediments were from the atomic bomb. However, more than 500 of atmospheric nuclear tests have been carried out worldwide since 1945, which caused global contamination of plutonium and other radionuclides. Table 1-1 shows the main radionuclides, which have long half-lives, released from the nuclear tests (UNSCEAR, 2000). Figure 1-2 represents the number of the tests in each year (UNSCEAR, 2000) and Fig.1-3 shows annual depositions of $^{239+240}$Pu and ^{137}Cs in Japan (Hirose et al, 2001). It is likely that the plutonium and ^{137}Cs from the nuclear tests fell around the reservoir and were added into the sediment. It makes difficult to identify the origin of plutonium and ^{137}Cs deposited in the sediments, because the information on the concentrations is not sufficient to identify the source.

The values of $^{239+240}$Pu/^{137}Cs, ^{241}Am/$^{239+240}$Pu and ^{238}Pu/$^{239+240}$ Pu activity ratios have been used to discriminate plutonium from the Nagasaki atomic explosion and that came from global fallout of the nuclear tests. However it has been pointed out that the $^{239+240}$Pu/^{137}Cs activity ratio is affected by the variation in soil composition and migration behavior of these elements in the soil over a long period of time (Mahara and Miyahara, 1984; Mahara and Kudo, 1995). The same thing can be said for ^{241}Am/$^{239+240}$Pu activity ratio.

In the case of the application of ^{238}Pu/$^{239+240}$Pu activity ratio, it is not necessary to consider difference of chemical behavior of ^{238}Pu and $^{239+240}$Pu. The ^{238}Pu/$^{239+240}$Pu activity ratio was reported to be 0.05 – 0.06 in soil at the Nishiyama area (Yamamoto et al., 1985), and 0.02 – 0.04 for global fallout released from the nuclear tests (Hardy et al., 1973). However the difficulty in distinguishing the difference of these activity ratios has been recognized, especially for the case of low plutonium concentration. Thus there is a limit to application of the ^{238}Pu/$^{239+240}$Pu activity ratio to the discrimination of plutonium origin.

Table 1-1 Main radionuclides produced and globally dispersed in atmospheric nuclear tests (UNSCEAR, 2000)

Nuclide	Half life (y)	Global release ($\times 10^{15}$ Bq)
^{3}H	12.33	186,000
^{14}C	5,730	213
^{90}Sr	28.78	622
^{137}Cs	30.07	948
^{239}Pu	24,100	6.52
^{240}Pu	6,563	4.35
^{241}Pu	14.35	142

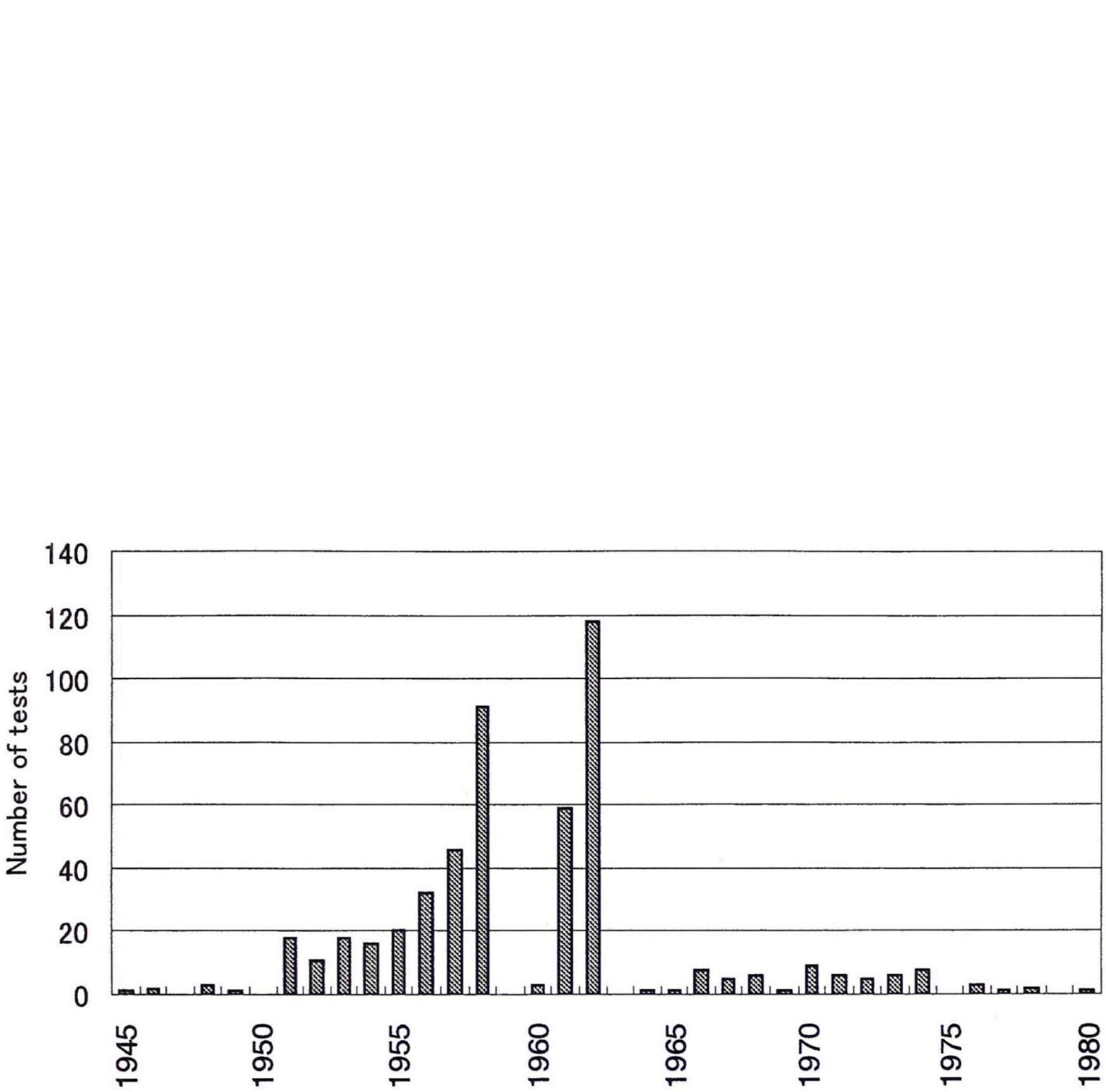

Fig. 1-2 Number of atmospheric nuclear tests from 1945 – 1980.
(UNSCEAR, 2000)

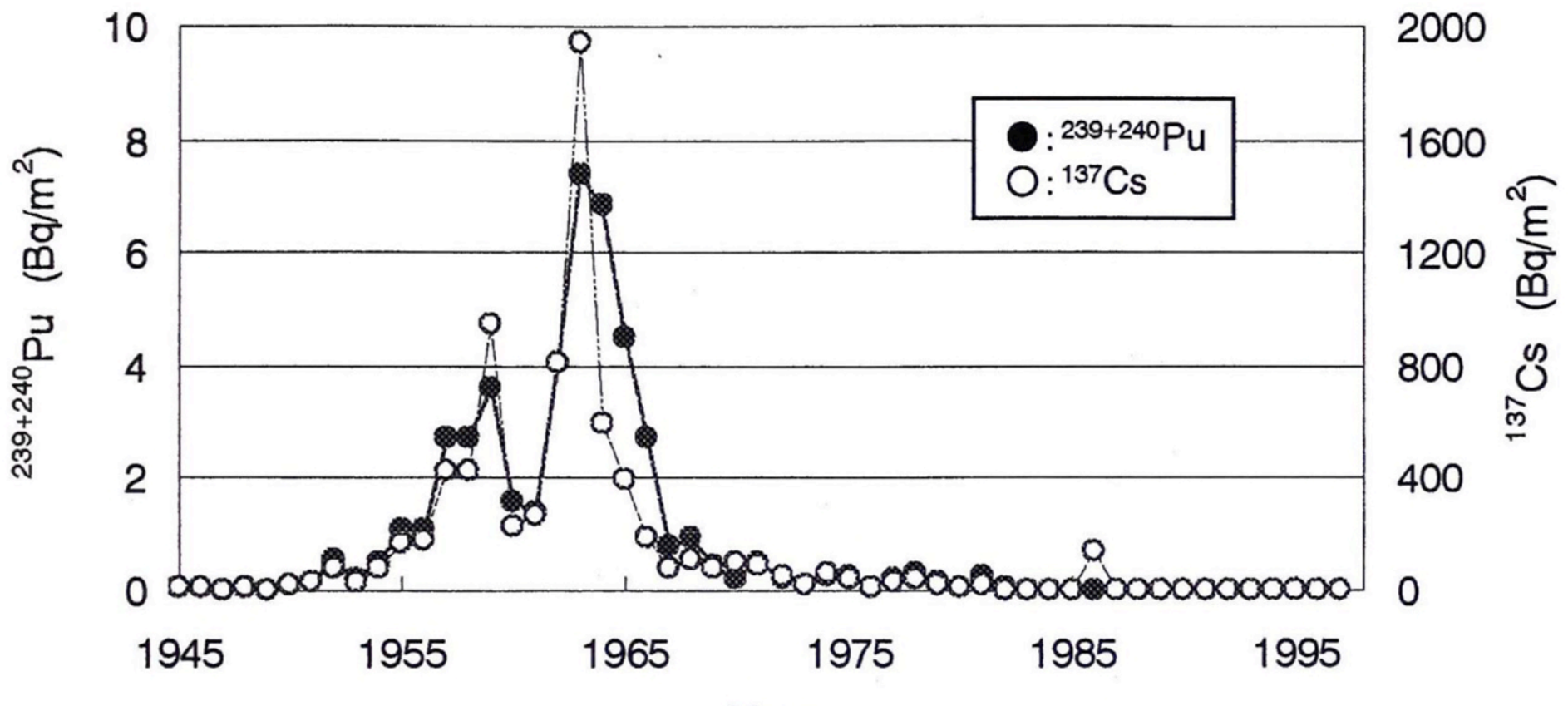

Fig. 1-3 Annual depositions of $^{239+240}Pu$ and ^{137}Cs observed at the Meteorogical Research Institute in Japan. (Hirose et al., 2001)

Isotopic ratio of $^{240}Pu/^{239}Pu$ has been recently used for the determination of the source of plutonium in environmental samples (e.g., Yamamoto et al., 1996; Boulyga et al., 2002). Plutonium-239 is source material of weapons and undergoes neutron capture reactions to produce ^{240}Pu during detonation of the weapons. The $^{240}Pu/^{239}Pu$ ratios in source materials are different according to the date when the weapons were made. It was reported that the isotopic ratios of the source materials before 1960 was less than 0.01, while those after 1960 are 0.06 (Rokop et al., 1996). The isotopic ratios in the fallout depends on the design of the weapons and the scale of explosion; the isotopic ratios during the nuclear tests ranged from 0.06 – 0.34 (Koide et al., 1985; Warneke et al., 2002). At present, the integrated ratio in the fallout is 0.176 ± 0.014 (Krey et al., 1976). Therefore, $^{240}Pu/^{239}Pu$ ratio is an effective tool for the identification of the source of plutonium.

In this work, sediment cores at the Nishiyama reservoir were collected and the depth profiles of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ and ^{137}Cs concentrations were determined in order to obtain temporal distribution of deposit of the plutonium from the atomic bomb in the sediments.

Spatial distribution of plutonium from the atomic bomb is another interest. Since deposition of plutonium retained about 97% of the total $^{239+240}Pu$ contents in soil of up to 30 cm depth (Mahara and Miyahara, 1984; Mahara and Kudo, 1995), the subject of this work was focused on surface distribution of the plutonium from the analysis of soil samples. Sakanoue and Tsuji (1971) studied $^{239+240}Pu$ concentrations in soil samples collected from northern, eastern and southern areas within 4 km of the hypocenter. They reported that samples taken from the Nishiyama area had the highest concentration. Following these findings, investigations were expanded over a wider area. Yamamoto et al. (1985) measured in soil samples collected from up to 30 km from the hypocenter and Kudo et al. (1991) studied soil samples collected within 100 km east and 3 km west from the hypocenter. In the results of these studies, the highest $^{239+240}Pu$ and ^{137}Cs concentrations were observed at the Nishiyama area and activity ratios of $^{239+240}Pu/^{137}Cs$, $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ were used to identify depositions from the

atomic bomb. Yamamoto et al. (1985) found that the plutonium from the atomic bomb was detectable at as far as 8 km east from the hypocenter and Kudo et al. (1991) reported that the plutonium concentration decreased to a background level at 18 km east from the hypocenter. To date, no extensive data has been reported on the $^{240}Pu/^{239}Pu$ ratio in the soil around the hypocenter at Nagasaki city, except three separate reports on soil samples taken from a limited area surrounding Nishiyama (Komura et al., 1984; Yamamoto et al., 1985; Muramatsu et al., 2003). The technique of $^{240}Pu/^{239}Pu$ ratio measurement was again applied to soils collected around the hypocenter in Nagasaki city and in eastern area of Nagasaki city to identify the distinct distribution of plutonium released from the atomic bomb.

2. Experimental

2.1 Sampling

2.1.1 Sediments

Sediment cores were collected at Nishiyama reservoir (Fig. 2-1). The reservoir with a dam was constructed in 1904 for water utilization at Nagasaki city. After 1982 when a heavy rainfall disaster (what is called 'Nagasaki daisuigai') was occurred, a new dam for flood control was constructed at about 60 m downstream from the old one. Table 2-1 shows an outline of the reservoir.

At the first sampling, a sediment core (herein after B core) was collected with a Mackereth type core sampler at the reservoir in December 1999. The sampling point of the core was about 370 m northeast from the old dam. The diameter of the core was 8 cm and the length was about 2.5 m.

At the second sampling in October 2005, a thin wall sampler (stationary piston type) was used to obtain a longer core compared with the B core. The core (herein after C core) was collected at about 80 m northeast from the old dam, which was located downstream from the point collected B core, because the influence of the disturbance by debris flow in the point collected the C core would be smaller than the upstream place. The sampling was conducted according to the standard of the Japan Geotechnical Society (JGS1221-2003). The diameter of the core was 8.6 cm and the length was about 6.2 m. As the sampler reached bedrock, the core was considered to contain all sediments that have been deposited since the construction.

2.1.2 Soils

Surface soils were collected in three area: (1) five samples (E2 – E6) collected around the Nishiyama reservoir in December 1999, (2) fifteen samples (E1, E7 – E13, N1, N2, W1 – W3, S1, S2) collected within a 10 km radius of the hypocenter in 2002 (Noritake et al., 2002) and (3) 41 samples (N3 – N6, K1 – K6, F1 – F3, S1 – S3, O1 – O6, P1 – P19) in the area covering the eastern part of Nagasaki prefecture, Saga prefecture, Fukuoka prefecture, Kumamoto

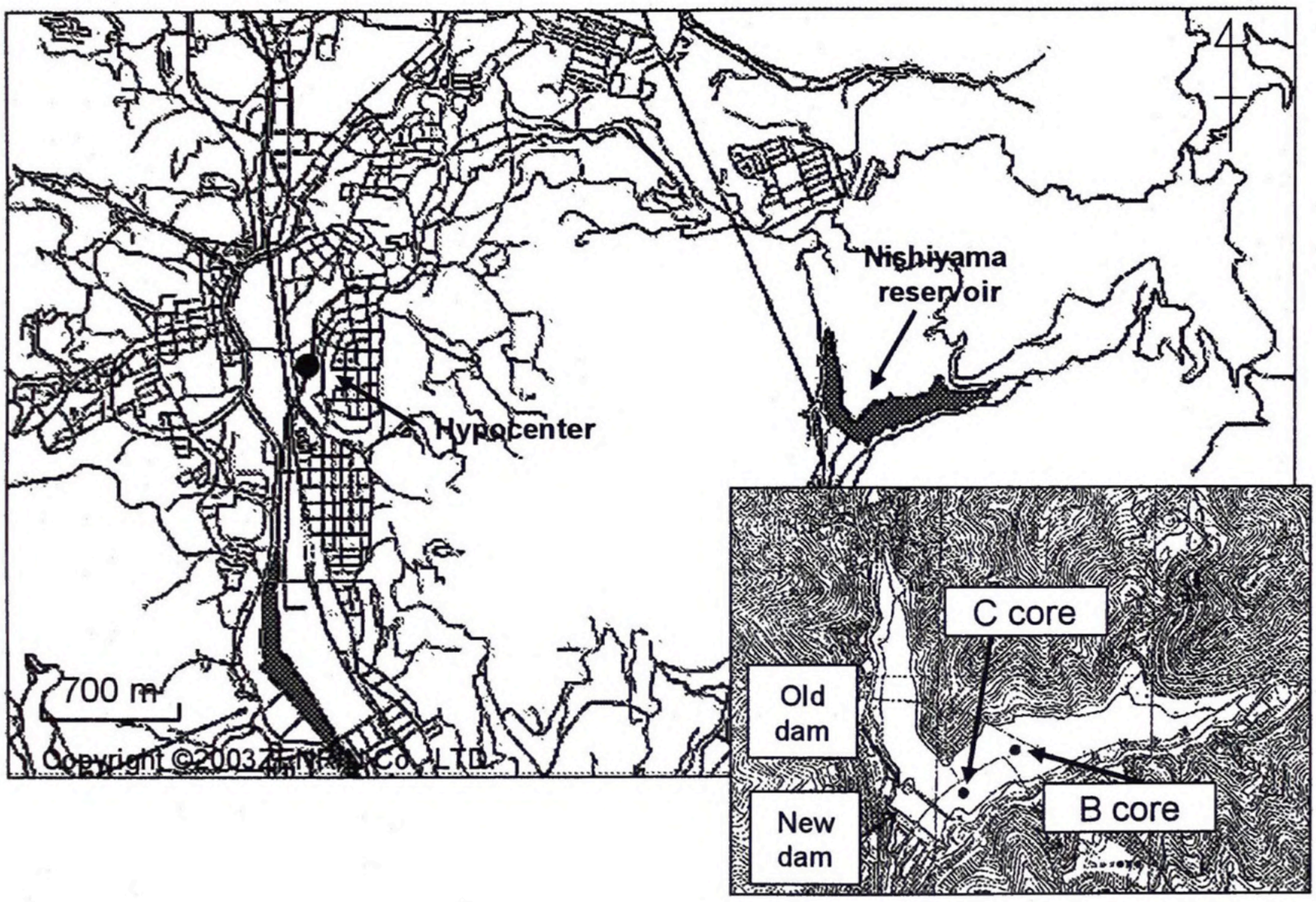

Fig. 2-1 Location of the Nishiyama reservoir and the sampling points of the sediment cores.

Table 2-1 Outline of Nishiyama reservoir

River/ Water system	Nishiyama river /Nakashima river system
Location	Katafuchi machi, Nagasaki city
Style of dam	Concrete gravity dam
Old dam (height/length/volume)	40 m/ 216 m/ 85,000 m^3
Catchment area	3.6 km^2
Reservoir area	0.15 km^2
Gross reservoir capacity	1,580,000 m^3
Active reservoir capacity	1,470,000 m^3
Irrigation capacity	760,000 m^3 (8,100 m^3/day)
Sediment capacity	110,000 m^3

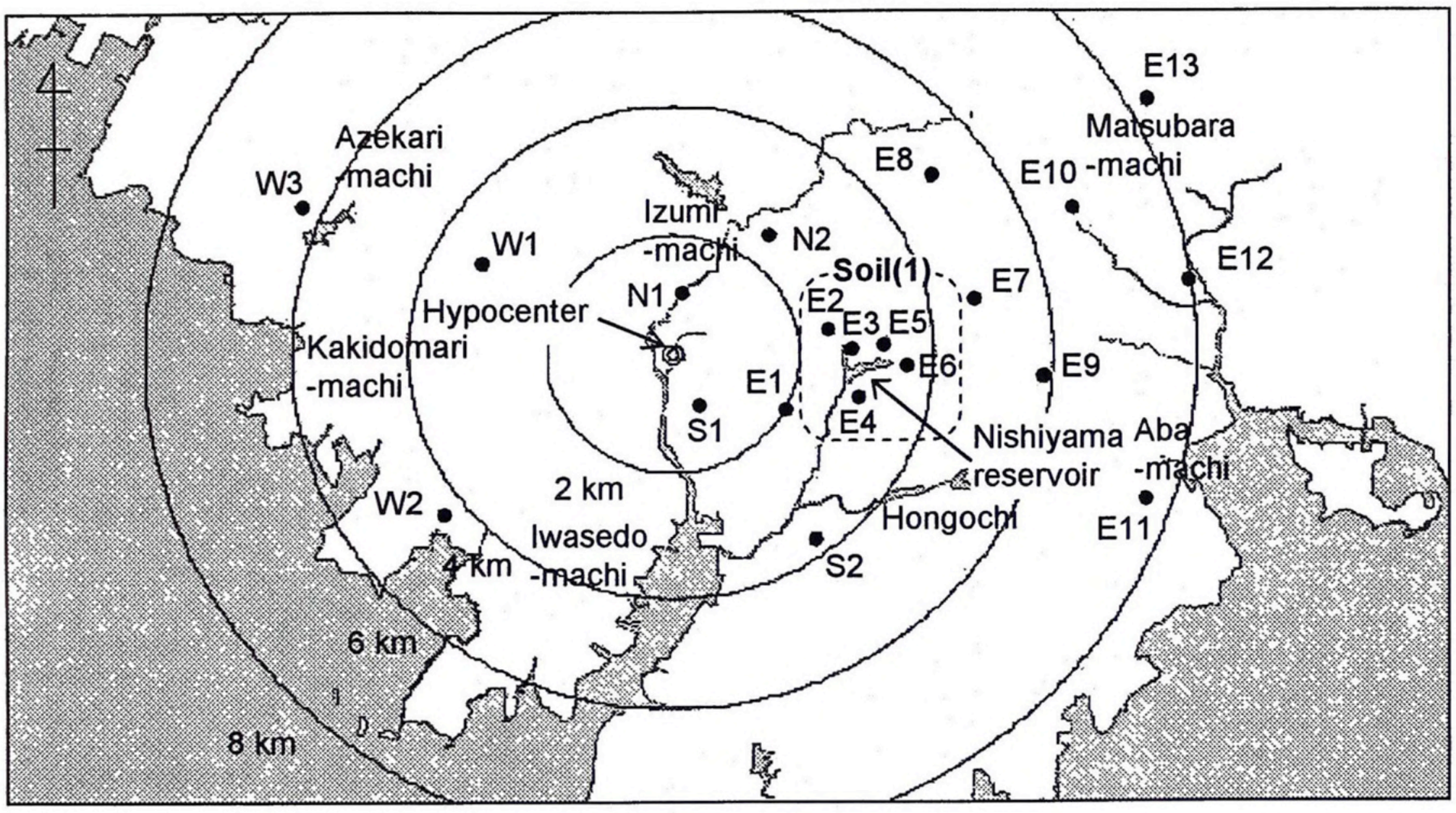

Fig. 2-2 Location of the sampling points of soil (1) and (2) within a 10 km radius of the hypocenter in Nagasaki city.

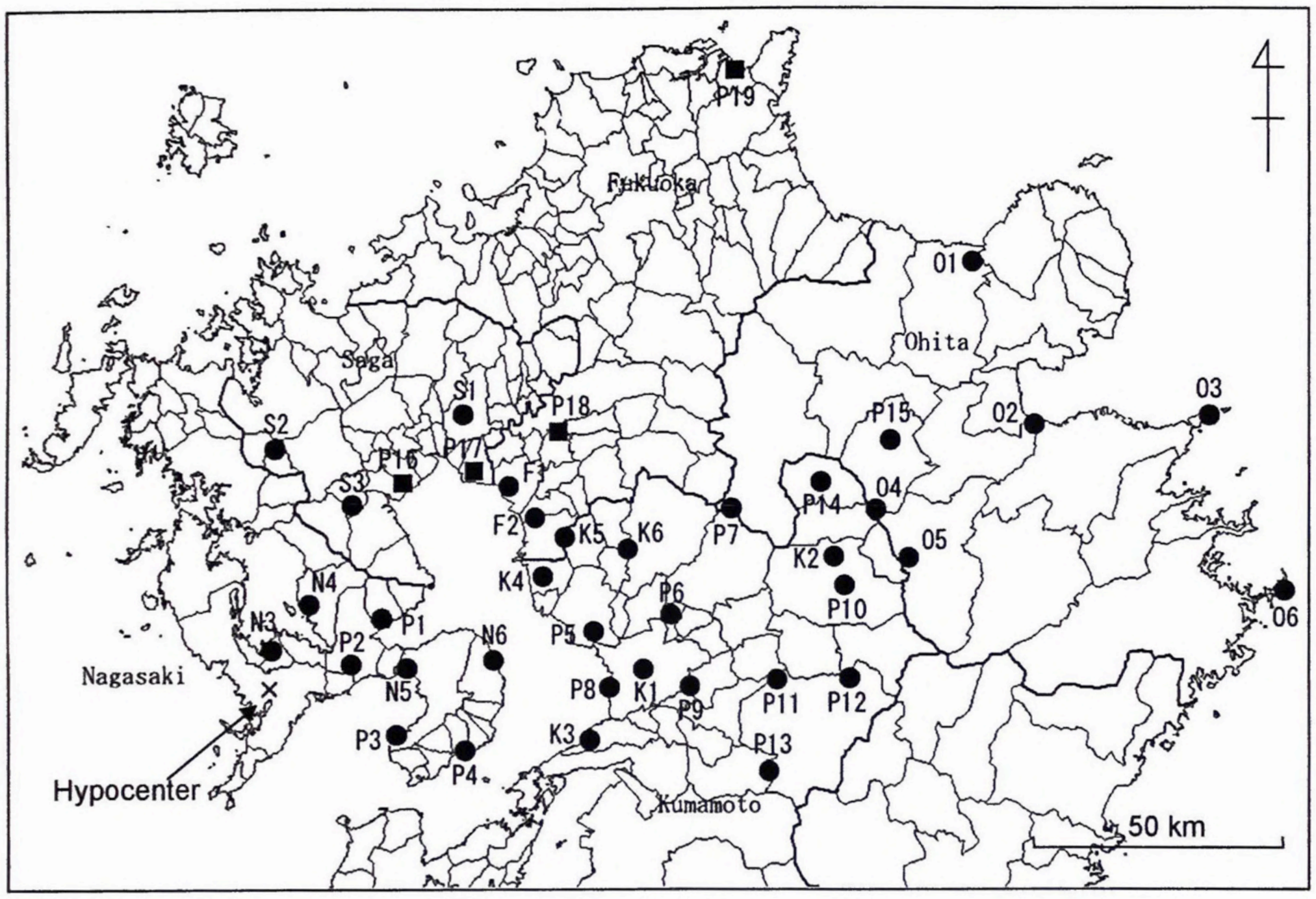

Fig. 2-3 Location of the sampling points of soil (3) in north part of Kyushu area.
(●: Uncultivated soil, ■: Agricultural soil)

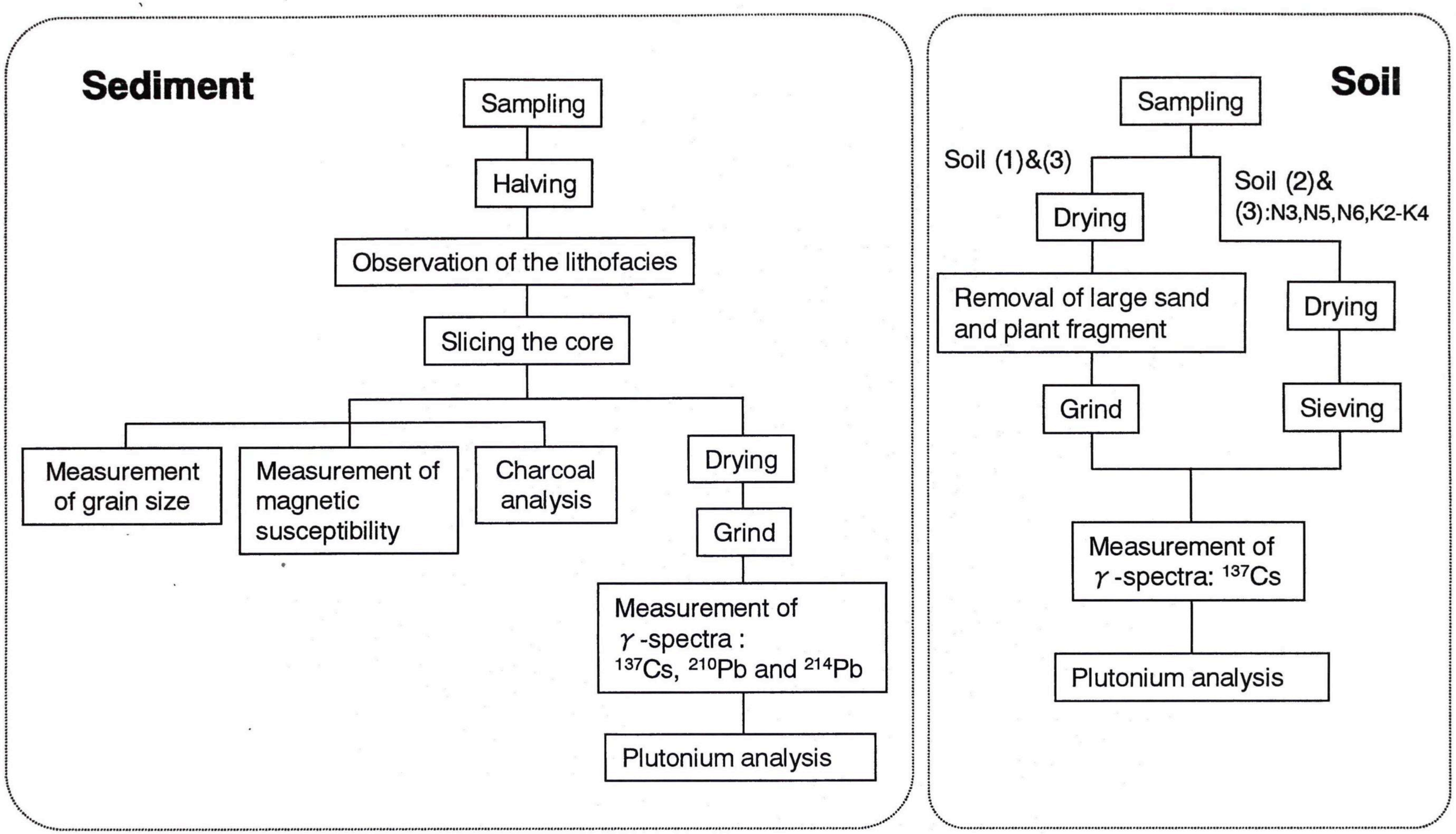

Fig. 2-4 Flow of the analytical procedure.

work. This method has high sensitivity and the operation of the instrument is easier than thermal ionization mass spectrometry which has been the primary method for the measurement of the isotopic ratio. Prior to ICP-MS measurements, it is necessary to separate plutonium from matrix of sediment and soil. This is because lead and uranium in the matrix form, with oxygen or hydrogen, polyatomic ions which have same mass number of plutonium isotopes (^{239}Pu: $^{207}Pb^{16}O_2$, $^{238}U^{1}H$, ^{240}Pu: $^{208}Pb^{16}O_2$), and because alkali elements cause to decrease the ionization efficiency of actinide isotopes (Magara et al., 2002).

2.2.1 Pretreatment and sediment characterization

(1) Sediments

First, the cores were halved vertically and the lithofacies were observed. The composition of sediments were mainly clay and silt with some laminated mud. There were some sand layers and plant fragments in the sediments. Photo 2-1 shows the cross section of the C core.

After the observation, the cores were sliced into samples of about 2 cm each in thickness. The samples were packed into plastic container and used to measure magnetic susceptibility with a Bartington MS2 meter (UK).

Grain size was analyzed with a Shimadzu SALD-3000S (Japan) by the following procedure. A portion of sample (about 0.5 g) was soaked in 5 ml of 6% H_2O_2 and placed in an oven for one hour at 110 'C to dissolve organic materials. Distilled water (20 ml) was added and placed over one day in the room temperature. After mixing, the suspension was added into 10% sodium hexametaphosphate solution in the instrument. An ultrasonic wave was irradiated for 30 seconds to disperse particles and then the grain size in sediment was measured.

Before plutonium analysis and measurement of γ-ray spectra, a half of the samples were dried and homogenized.

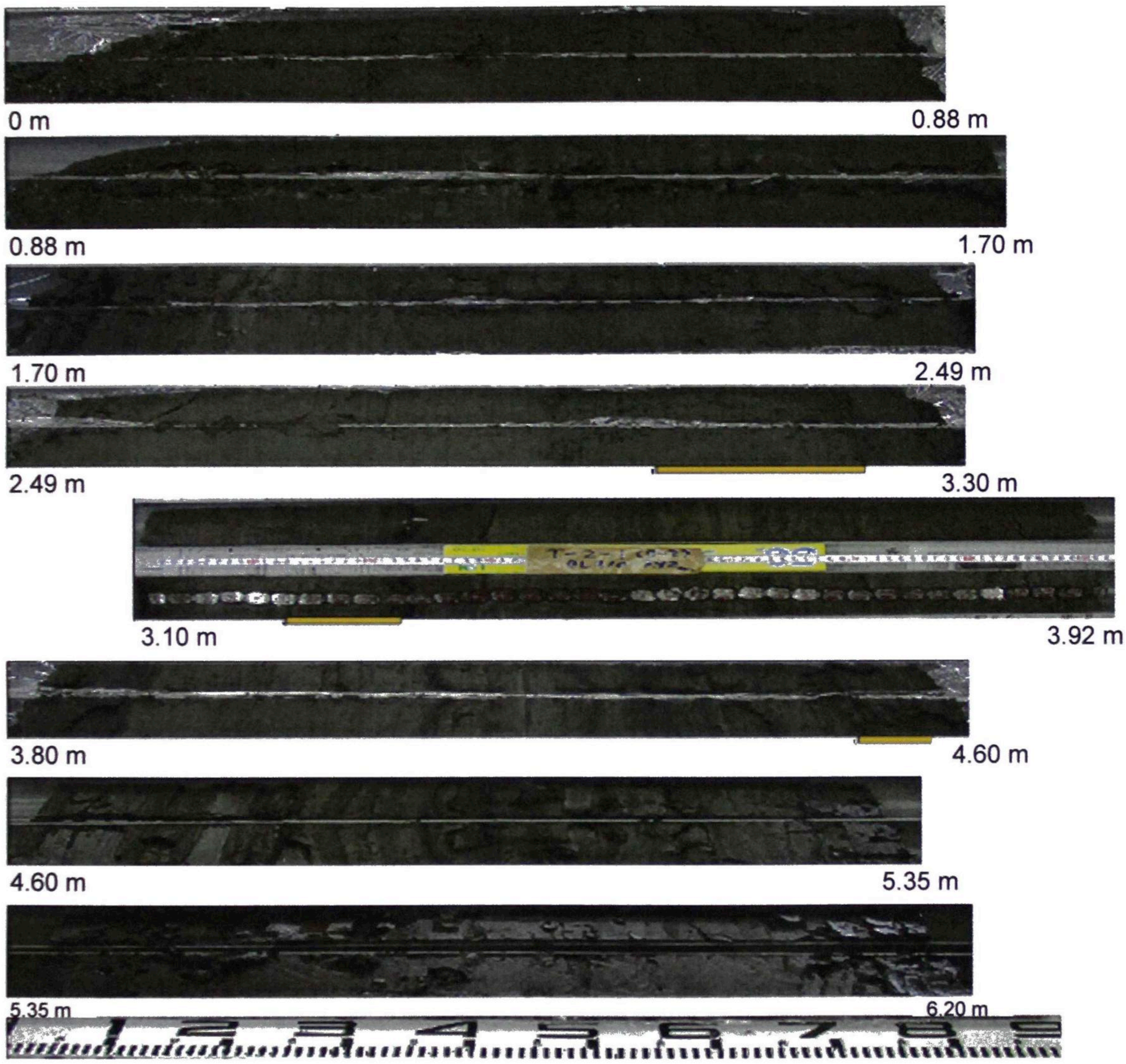

Plate 2-1 Photograph of cross section of the C core.

(2) Soils

Soil (1) and (3): After drying until constant weight at 70 ˚C, large sand particles and fragments of plants were removed. These soils then were ground with a mortar and pestle. For the samples of N3, N5, N6 and K2 – K4, sieving was applied.

Soil (2): After drying at 110 ˚C for 24 hours, these samples were sieved through a mesh (mesh size: 2 mm).

2.2.2 Plutonium analysis

(1) Chemical separation of plutonium

The chemical separation and the measurement by ICP-MS were operated in clean rooms (ISO class 5 and 6, respectively) in Japan Atomic Energy Agency. These rooms have the ability to measure the isotopic ratio of trace amounts of uranium and plutonium in environmental samples (Magara et al., 2000). The procedure used for plutonium leaching was based on the method reported by Muramatsu et al. (1999). Figure 2-5 shows the scheme of the chemical separation.

The dry sample (about 1 – 2 g) was taken into a beaker, together with 4 pg of ^{242}Pu (CRM130, Plutonium Spike Assay and Isotopic Standard, New Brunswick Laboratory, US) as a spike for the isotope dilution method. Then 8M HNO_3 (quintuple weight to the sample) was added and the mixture solution was heated on a hot plate to leach plutonium. After 3 hours, the solution was centrifuged. The residue was boiled again with 8M HNO_3 of twice the sample weight. This leaching process was repeated twice. The supernatant resulting from the three steps of leaching was dried up and the obtained paste was dissolved in 3 ml of 8M HNO_3, which was followed by filtration through a Teflon membrane filter (pore size: 0.45 μm, DISMIC, Advantec, Japan) to remove residue. The beaker was rinsed twice with 1 ml of 8M HNO_3, which was also filtered. All the three filtrates were mixed and sodium nitrite (200 mg) was added. The solution was left overnight to adjust plutonium to the tetravalent state.

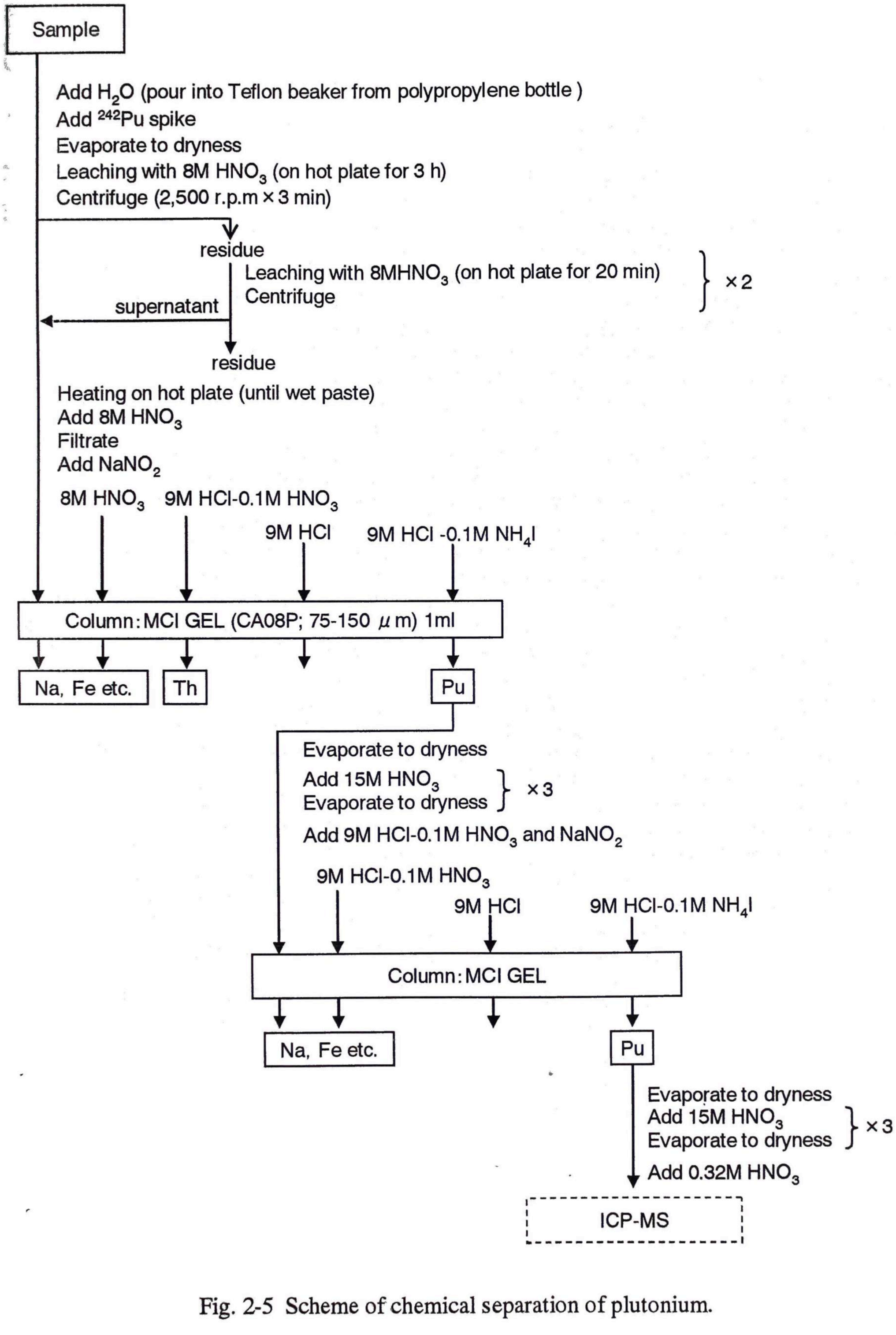

Fig. 2-5 Scheme of chemical separation of plutonium.

Plutonium purification was carried out with a 1 ml column containing anion-exchange resin (75 – 150 μm, MCI GEL CA08P, Mitsubishi Chemical, Japan). After resin was pretreated with 8M HNO_3 to be NO_3^- form, plutonium in the tetravalent state was loaded onto the column. The column was rinsed sequentially with 8M HNO_3 (2 ml), 9M HCl - 0.1M HNO_3 (7 ml) and then 9M HCl (5 ml). Plutonium was stripped from the column as Pu(III) with 3 ml of 9M HCl - 0.1M NH_4I. Then, the column was left for 3 hours to complete the reduction to Pu(III) and another 4 ml of the elutant was passed through the column.

In order to decompose organic material and to expel iodine completely from the collected fraction, the solution was heated at 200' C on a hot plate, and then the solution, to which 15M HNO_3 was added was heated again until the residue became colorless. The residue was dissolved in 3 ml of 9M HCl - 0.1M HNO_3 and 200 mg of sodium nitrite was added. The solution was left overnight for plutonium valence adjustment, and then second purification was carried out in the same manner mentioned above, except the rinsing step; 9M HCl - 0.1M HNO_3 (4 ml) and 9M HCl (5 ml). Finally, the residue was dissolved in 2 – 3 ml of 0.32M HNO_3, a solution suitable for ICP-MS measurement.

(2) Determination of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ concentration by ICP-MS

The instrument used was a double-focusing ICP-MS (ELEMENT1, Thermo Fisher Scientific, US) with Guard Electrode, operated in electrostatic scanning mode (E-scan) with a resolution (M/ΔM) of 300. A micro flow nebulizer made of tetra fluoro ethylene-perfluoro alkylvinyl ether copolymer (PFA) was used as a sample introduction system. The plutonium isotopic standard (NIST SRM 947) was used to correct the mass bias of the instrument. Since polyatomic ion of ^{238}U with H interferes with the measurement of ^{239}Pu, preliminarily obtained ratio of $^{238}UH/^{238}U$ (4×10^{-5}) was used to correct this interference. Concentrations of ^{239}Pu and ^{240}Pu were obtained from isotopic dilution method by using a ^{242}Pu spike. The data of the concentration were expressed by the sum of ^{239}Pu and ^{240}Pu concentrations in terms of unit of radioactivity for the convenience to compare the data with those obtained with α-ray

spectrometry in other studies. Details of the operating conditions and the method of mass bias correction were reported by Magara et al. (2002). Typical operating conditions for the ICP-MS are summarized in Table 2-2.

2.2.3 Other analyses

(1) ^{137}Cs, ^{210}Pb and ^{214}Pb

The sediment samples (about 20 g) were analyzed for their contents of ^{137}Cs, ^{210}Pb and ^{214}Pb by using high-purity (HP) Ge detector (EGNC80-230R, EURISYS MESURE, France or LOAX60450/30P, ORTEC, US). The activities were decay-corrected to the sampling date. In order to estimate sedimentation rate by using ^{210}Pb dating method, activity of excess ^{210}Pb was calculated from subtracting ^{214}Pb activity from ^{210}Pb one assuming that activity of ^{214}Pb was equal to that of ^{226}Ra.

The activities of ^{137}Cs in the soil (1) and (3) (about 1 – 2 g) were measured with HP-Ge detector (GWL-220230-S, ORTEC, US). The measured activity of ^{137}Cs in the samples was decay-corrected to those in July 2002, the date when the samples of soil (2) were collected.

In the measurement of the sediments and the soils, the counting efficiency was calibrated from measuring two standard reference materials (NIST SRM4350B: River sediment, SRM 4354: Freshwater lake sediment).

(2) Charcoals

The analyses of macroscopic and microscopic charcoals were based on the method reported by Yasuda (2004). The samples were the sediments at the depth between 300 and 550 cm in the C core. The wet sample of 1 cm^3 was immersed one night in a solution of 5%(w/w) of sodium hexametaphosphate to disperse the sediment aggregates and to facilitate sieving. Markers (Microsphere, 15 μm, DuPont, US) were added to compare the microscopic charcoal. The suspension was sieved through a mesh (mesh size: 125 μm). The macroscopic charcoals in the residue on the mesh were counted under a loupe (Steinheil 10×, Peak, Japan). The

precipitation in the filtrate was soaked in warm HF solution (46 – 48 %) to destroy silica. Glycerin jelly was added to the residue and the suspension containing microscopic charcoal was dropped on to slide glass. Microscopic charcoals were counted under a microscope (DFC320, Leica, Germany).

Table 2-2 Typical operating conditions for ICP-MS

RF power	1148 W
Cooling gas flow rate	16.0 l/min
Auxiliary gas flow	0.85 l/min
Sample gas flow	1.010 - 1.060 l/min
Solution uptake rate	About 0.2 ml/min
Mass window	5%
Measured mass	238, 239, 240, 242
Sampling time	50 ms
Scan per replicate	400 times
Replicate	5 times
Detector dead time	18 ns

3. Temporal distribution of plutonium from Nagasaki atomic bomb deposited in sediment of Nishiyama reservoir

Depth profiles of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ and ^{137}Cs concentrations in the B and C cores were determined in order to obtain temporal distribution of plutonium and ^{137}Cs deposited in sediments for 60 years. Figure 3-1 shows depth profiles of the $^{240}Pu/^{239}Pu$ ratio, the $^{239+240}Pu$ and ^{137}Cs concentrations in the B core collected at the first sampling. The $^{239+240}Pu$ and ^{137}Cs concentrations were detected in a sediment at bottom of the core, which indicated that the sediment was deposited after 1945 (The details were described in Chap. 3.1). It did not permit us to understand the temporal distribution immediately after the detonation of the atomic bomb. On the contrary, the C core collected at the second sampling contained sediments that have been deposited since the reservoir was constructed, which enable us to obtain entirety of the temporal distribution. Therefore, the results of the C core were discussed predominantly in comparison with those in the B core. The $^{240}Pu/^{239}Pu$ ratio, the $^{239+240}Pu$, ^{137}Cs and excess ^{210}Pb concentrations in the core are shown in Table 3-1, and the depth profiles are shown in Fig. 3-2.

3.1 Identification of chronological markers of sediments for deposits in 1945 and 1963

The maximum $^{239+240}Pu$ and ^{137}Cs concentrations in the C core were observed in a sediment at the depth of 435 – 437 cm. The $^{240}Pu/^{239}Pu$ ratio in the sediment was 0.0283. This value was considerably lower than the global fallout value of the nuclear tests (0.176 ± 0.014) (Krey et al., 1976). It was reported by Rokop et al. (1996) that the isotopic ratio of the source material of atomic bombs before 1960 was less than 0.01. Since the plutonium used for the Nagasaki atomic bomb was made before 1960, its isotopic ratio was likely to be the same level. However, the obtained isotopic ratio was higher than that in the source material. This is probably because neutron capture reactions at the explosion increased the isotopic ratio. In the case of nuclear tests which are the main origin of global fallout, the isotopic ratio of the source material was higher (about 0.06) (Rokop et al., 1996) and the scale of explosion was bigger resulting in the sufficiently higher isotopic ratios of nuclear tests fallout (0.06 –

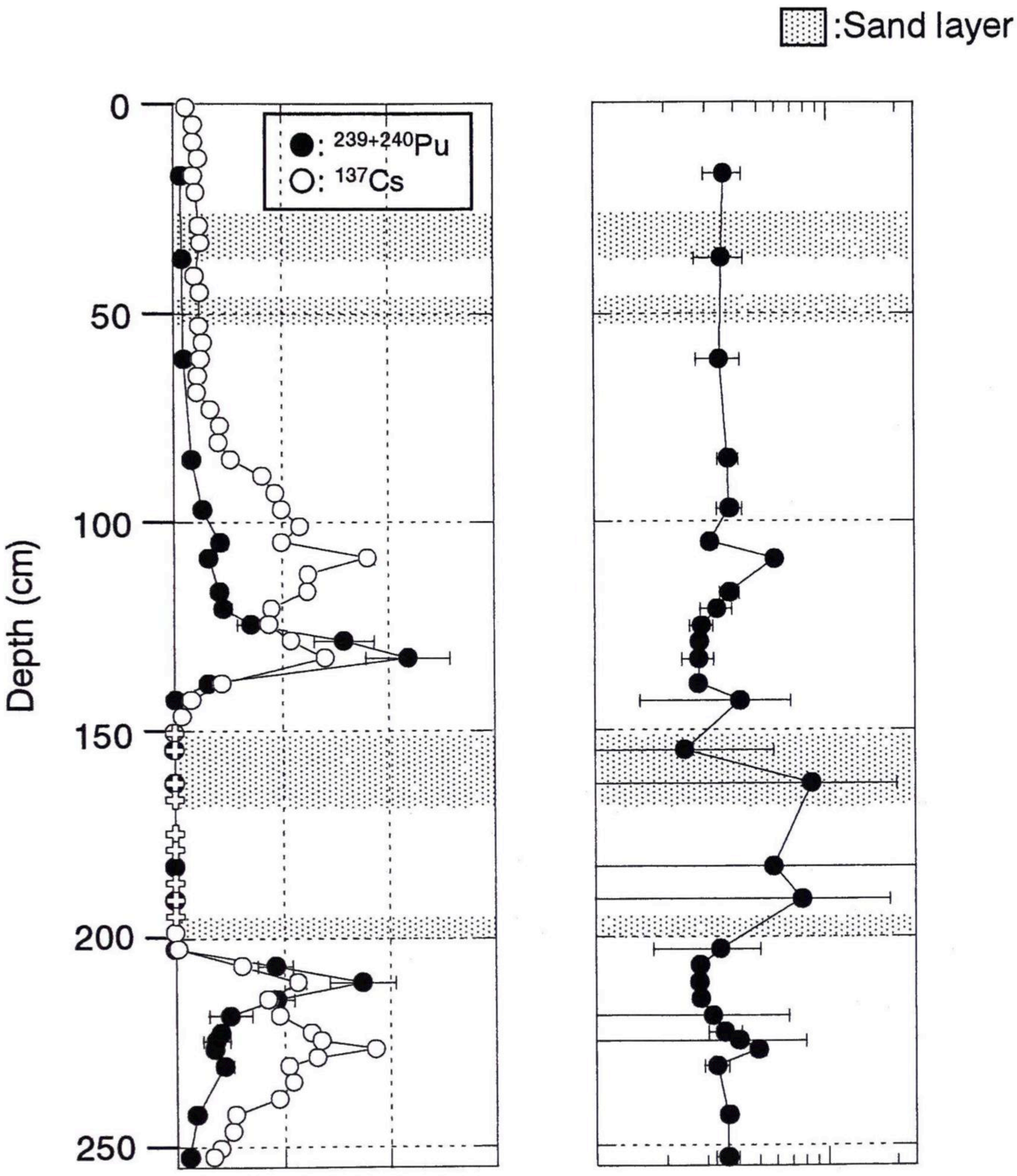

Fig. 3-1 Depth profiles of $^{239+240}Pu$ and ^{137}Cs concentrations and $^{240}Pu/^{239}Pu$ ratio in the B core.

* Mark ✪ indicated that the ^{137}Cs concentration was under the detection limit.

Table 3-1 Isotopic ratio of $^{240}Pu/^{239}Pu$ and concentrations of $^{239+240}Pu$, ^{137}Cs and excess ^{210}Pb in the C core

(Error :2SD)

Sample ID	Depth (cm)	$^{240}Pu/^{239}Pu$ ratio	$^{239+240}Pu$ (Bq/kg)	^{137}Cs (Bq/kg)	Excess ^{210}Pb (Bq/kg)
T-1-1	0 - 2	0.034 ± 0.003	4.5 ± 0.8	14 ± 2	269 ± 21
T-1-3	5 - 7	0.034 ± 0.004	3.7 ± 0.6	16 ± 3	269 ± 23
T-1-10	21 - 23	0.033 ± 0.002	4.1 ± 0.7	12 ± 2	188 ± 16
T-1-20	44 - 46	0.033 ± 0.003	4.9 ± 0.9	12 ± 1	67 ± 9
T-1-30	67 - 69	0.034 ± 0.002	4.5 ± 0.8	13 ± 2	107 ± 12
T-2-6	100 - 102	0.034 ± 0.004	3.9 ± 0.7	15 ± 2	116 ± 12
T-2-16	123 - 125	0.033 ± 0.004	4.0 ± 0.7	14 ± 2	91 ± 11
T-2-27	148 - 150	0.034 ± 0.005	4.1 ± 0.7	15 ± 1	73 ± 9
T-3-2	172 - 175	0.033 ± 0.003	3.5 ± 0.6	10 ± 2	53 ± 10
T-3-13	198 - 200	0.034 ± 0.002	4.7 ± 0.8	11.2 ± 0.9	18 ± 5
T-3-25	225 - 228	0.034 ± 0.003	5.1 ± 0.9	10 ± 2	9 ± 6
T-4-2	252 - 254	0.035 ± 0.002	4.4 ± 0.8	12 ± 2	27 ± 9
T-4-11	275 - 278	0.034 ± 0.003	5 ± 1	10 ± 2	14 ± 8
T-4-20	298 - 301	0.033 ± 0.002	3.3 ± 0.6	9 ± 2	9 ± 6
T-2-1-17	327 - 329	0.038 ± 0.004	4.6 ± 0.8	18 ± 2	84 ± 11
T-2-1-22	338 - 340	0.038 ± 0.003	4.6 ± 0.8	22 ± 2	56 ± 11
T-2-1-28	351 - 353	0.041 ± 0.001	8 ± 1	41 ± 4	67 ± 14
T-2-1-31	358 - 360	0.040 ± 0.003	9 ± 2	47 ± 3	63 ± 13
T-2-1-33	362 - 364	0.042 ± 0.004	11 ± 2	57 ± 4	83 ± 15
T-2-1-34	364 - 366	0.037 ± 0.001	14 ± 2	49 ± 2	17 ± 6
T-2-1-35	366 - 369	0.061 ± 0.003	13 ± 2	88 ± 3	43 ± 10
T-2-1-36	369 - 373	0.066 ± 0.005	14 ± 2	124 ± 7	88 ± 20
T-5-1	380 - 382	0.046 ± 0.002	14 ± 2	77 ± 7	83 ± 21
T-5-2	382 - 384	0.042 ± 0.002	14 ± 2	52 ± 3	4 ± 7
T-5-5	389 - 391	0.036 ± 0.001	17 ± 3	-	-
T-5-11	402 - 404	0.029 ± 0.002	31 ± 5	31 ± 2	-
T-5-15	411 - 413	0.0282 ± 0.0006	43 ± 8	30 ± 3	4 ± 6
T-5-17	415 - 417	0.028 ± 0.006	49 ± 11	-	-
T-5-20	422 - 424	0.028 ± 0.001	101 ± 18	-	-
T-5-23	428 - 431	0.028 ± 0.003	157 ± 30	-	-
T-5-25	433 - 435	0.0283 ± 0.0003	230 ± 40	83 ± 6	-
T-5-26	435 - 437	0.0283 ± 0.0004	303 ± 53	130 ± 4	20 ± 7
T-5-27	437 - 439	0.0285 ± 0.0005	241 ± 42	-	-
T-5-28	439 - 442	0.028 ± 0.003	266 ± 49	114 ± 3	17 ± 7
T-5-29	442 - 444	0.0285 ± 0.0009	30 ± 5	20 ± 3	-
T-5-31	446 - 448	n.d.	0.029 ± 0.006	5 ± 1	8 ± 5
T-5-34	453 - 455	n.d.	< 0.003	0.7 ± 1	14 ± 5
T-6-2	462 - 464	0.046 ± 0.042	0.10 ± 0.03	< 0.07	18 ± 6
T-6-3	464 - 467	n.d.	0.031 ± 0.009	-	-
T-6-4	467 - 469	0.07 ± 0.04	0.028 ± 0.008	-	-
T-6-9	478 - 480	0.042 ± 0.035	0.04 ± 0.01	-	-
T-6-10	480 - 482	0.03 ± 0.01	0.06 ± 0.01	< 0.06	11 ± 5
T-6-11	482 - 484	n.d.	< 0.007	-	-
T-6-20	502 - 504	n.d.	< 0.0039	-	-
T-6-30	524 - 526	n.d.	< 0.0026	-	-
T-7-5	545 - 547	0.19 ± 0.03	0.07 ± 0.01	< 0.1	19 ± 8
T-7-6	547 - 549	n.d.	0.03 ± 0.01	-	-
T-7-7	549 - 552	0.03 ± 0.02	0.06 ± 0.01	-	-
T-7-15	569 - 571	n.d.	< 0.0024	< 0.1	17 ± 8
T-7-25	593 - 595	n.d.	< 0.0038	< 0.1	18 ± 9
T-7-35	617 - 619	0.03 ± 0.01	0.08 ± 0.02	21 ± 2	2 ± 8

n.d.: not detectable.

-: no data.

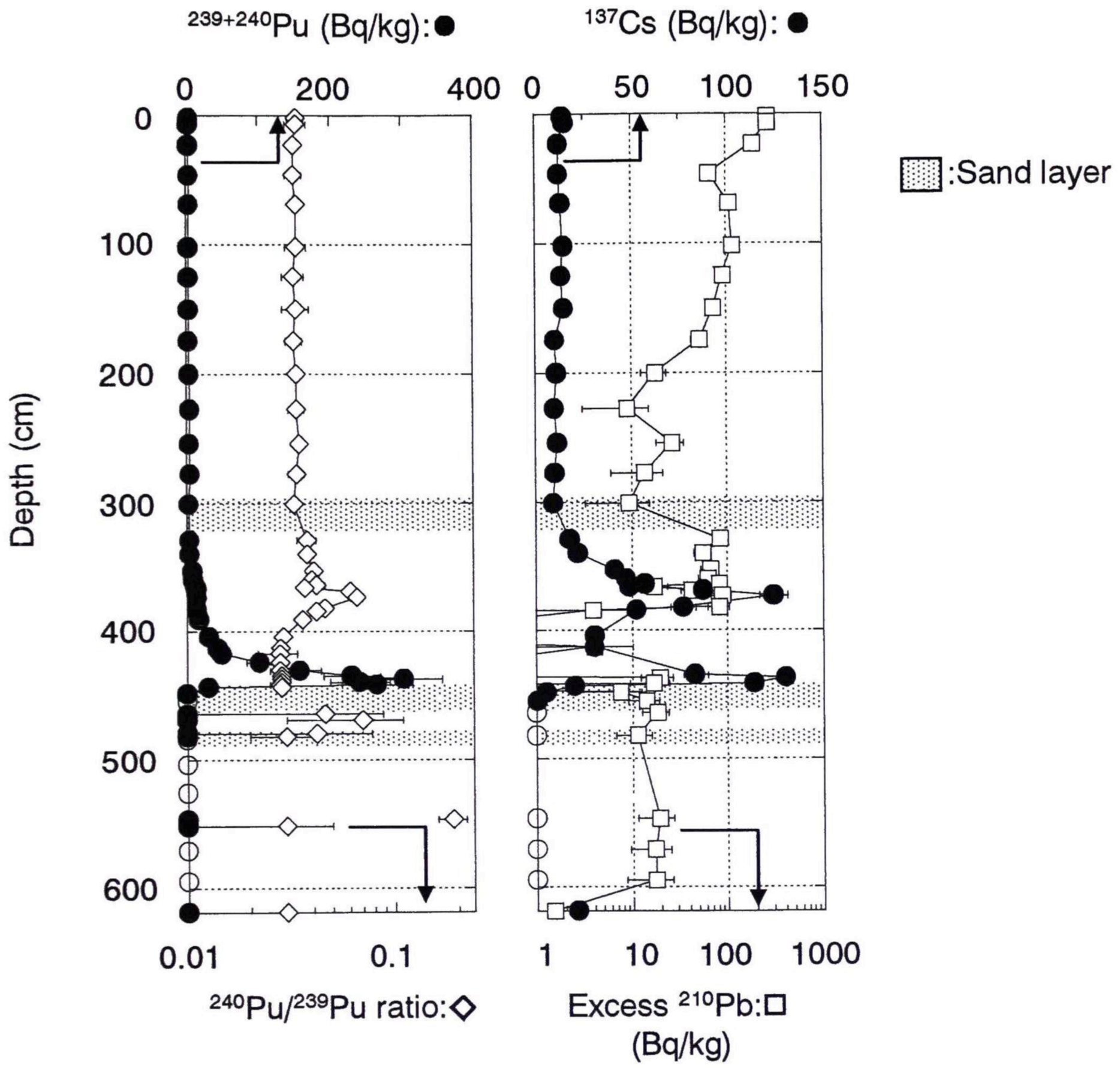

Fig. 3-2 Depth profiles of $^{239+240}Pu$ concentration and $^{240}Pu/^{239}Pu$ ratio, ^{137}Cs and excess ^{210}Pb concentrations in the C core.

* Each open circle indicated that the $^{239+240}Pu$ and ^{137}Cs concentrations were under the detection limit.

0.34) (Koide et al., 1985; Warneke et al., 2002). At the depth below 450 cm, the concentrations in sediments were very low or less than detection limit. It is reasonable that these low values were due to the cross contamination when the core was sampled or sliced. It is, therefore, deduced that the sediments were deposited before 1945, the period when no artificial radionuclides had been released into the world. From these results, the plutonium and ^{137}Cs deposited in the sediment at the depth of 435 – 437 cm were attributed only to the Nagasaki atomic bomb and the peaks were formed at 1945 by deposition after the detonation.

Another peak of ^{137}Cs concentration and a peak of $^{240}Pu/^{239}Pu$ ratio were found in a sediment at the depth of 369 – 373 cm, while the $^{239+240}Pu$ concentrations in sediments (364 – 384 cm) were almost constant. It was known that global release of ^{137}Cs and $^{239+240}Pu$ were 948×10^{15} Bq and 10.87×10^{15} Bq, respectively (UNSCEAR, 2000; see Table 1-1), resulting in that the inventory of ^{137}Cs was about 100 times higher than that of $^{239+240}Pu$. It was also reported that although the fallout of $^{239+240}Pu$ and ^{137}Cs had increased since 1945, the maximum depositions in Japan were observed in 1963 and then those had decreased (Hirose et al., 1986, 2001; see Fig. 1-3). Furthermore the isotopic ratio found in deposition of 1962 – 1964 was 0.16 – 0.22 (Koide et al., 1985; Warneke et al., 2002), which was higher than the value found in the sediment (435 – 437 cm) containing the plutonium only from the atomic bomb. From these, the observed profiles reflected atmospheric nuclear tests and the peaks at the depth of 369 – 373 cm corresponded to the maximum depositions in 1963.

Consequently, these peaks at the depth of 435 – 437 cm and 369 – 373 cm in the core were related to two depositional events occurred at 1945 and 1963, respectively, and the sediment observed the peaks become chronological markers in the core.

On the other hand, the depth profiles in the B core were not similar to those in the C core. Two sediments at the depth of 132 – 134 cm and 210 – 212 cm had peaks of the $^{239+240}Pu$ and ^{137}Cs concentrations and low $^{240}Pu/^{239}Pu$ ratios, and other two sediments at the depth of 108 – 110 cm and 226 – 228 cm had peaks of the $^{240}Pu/^{239}Pu$ ratio and the ^{137}Cs concentration. The $^{240}Pu/^{239}Pu$ ratio in sediment at the bottom of the core was equal to those in the surface

sediments in the B and C cores and higher than the value in sediment which contain he plutonium from the atomic bomb. It indicated that source of the plutonium in the sediment was the atomic bomb and the nuclear tests and that the B core was insufficient to understand the depositional records since the atomic bomb exploded.

3.2 Examination of depositional records of plutonium and ^{137}Cs for the last 60 years

In the C core, the $^{239+240}Pu$ concentration in sediments above 450 cm increased abruptly and the peak was formed. In sediments at the depth of 402 – 435 cm, the concentration decreased gradually. The concentration profile hold a record of the deposit from the atomic bomb after the detonation; the plutonium released by the detonation of the atomic bomb was deposited rapidly and then that deposited around the reservoir was carried and deposited in the sediments.

The $^{240}Pu/^{239}Pu$ ratio in the sediments increased and then decreased with shallowing the depth from 391 cm. This was caused by deposition of plutonium from the atmospheric nuclear tests. However it is interesting to note that the isotopic ratios indicate that the plutonium from the atomic bomb was dominant in deposit. The contribution of the atomic bomb to total deposit of plutonium in the sediment at the depth of 369 – 373 cm was estimated. The author assumed that isotopic ratio of the atomic bomb was 0.0283 ± 0.0002, which was the average of the sediments at the depth of 402 – 448 cm while the isotopic ratio in global fallout of the nuclear tests was 0.16 – 0.22 (Koide et al., 1985; Warneke et al., 2002). The fraction of the atomic bomb calculated was 67 – 83%.

In sediments between the surface and 301 cm, the $^{240}Pu/^{239}Pu$ ratios and the $^{239+240}Pu$ concentration were nearly constant. The averages of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ concentration were 0.0337 ± 0.0006 and 4.3 ± 0.6 Bq/kg, respectively. The profiles were similar to those in the B core. Moreover the isotopic ratios were comparable to those (0.032 – 0.039) in soil (1) collected around the reservoir (samples of E2 – E6, see Chap. 4.1) and the value (0.02 – 0.042) reported in the previous studies for soils collected at the Nishiyama area

(Komura et al., 1984; Yamamoto et al., 1985; Muramatsu et al., 2003). It is verified that the mixture of plutonium from the atomic bomb and the nuclear tests has been deposited in the sediments and that source of the supply is soils around the reservoir.

By using the estimation mentioned above, the fraction of the atomic bomb in the sediments between the surface and 301 cm was 96.3 ± 0.5 % under the assumption that $^{240}Pu/^{239}Pu$ ratio of present global fallout was 0.176 ± 0.014 (Krey et al., 1976). This is comparable to the reported values of 95 – 97% calculated from $^{240}Pu/^{239}Pu$ ratio measured by Lx/α-ray (Yamamoto et al., 1983). These results indicated that the fraction of the atomic bomb in sediments deposited recently was larger than that in the period when nuclear tests were conducted. This is due to decrease of deposition of the nuclear tests (see Fig. 1-3).

Annual flux of the plutonium from the atomic bomb into the reservoir in recent years was calculated from the results in this study and data of sedimentation reported by Nagasaki prefecture (2006). The flux was obtained by multiplying the sedimentation volume, $^{239+240}Pu$ concentration in surface sediment and the fraction of the atomic bomb (96.3%) together. The average of plutonium from the atomic bomb during last 6 years was 13 ± 4 MBq/y (6 ± 2 mg/y). In order to estimate the fluxes before 2000, determination of sedimentation rate was attempted by using ^{210}Pb dating, however, reliable data could not be obtained because the depth profile of excess ^{210}Pb concentration cannot fit to the decay curve (Fig. 3-2).

On the other hand, the depth profile of ^{137}Cs concentration had two peaks. Each peak reflected the depositions of the atomic bomb and the nuclear tests. Above 301 cm, the concentrations were almost constant, 12 ± 2 Bq/kg, which presumed that the ^{137}Cs derived from both sources continue to flow into the reservoir. Considering the origins and the quantities of release, temporal distributions of ^{137}Cs and $^{239+240}Pu$ concentrations were consistent. Although the ^{137}Cs concentration itself does not have information of the source, the depth profile of the ^{137}Cs concentration is complementary to that of plutonium and combination of both results of the plutonium and ^{137}Cs profiles might enable us to understand reliable depositional trend of plutonium released from the atomic bomb and the nuclear tests.

3.3 Estimation of other events recorded in the sediment core

The results of the sedimentology and charcoals concentrations in the C core were shown in Fig. 3-3. Three sandy layers were observed with increases in the profile of the magnetic susceptibility. It is known that an interbeded layer of a large volume of sand is caused by flood or earthquake (Arnaud et al., 2002, 2006; Nomada et al., 2005). Chronological markers indicated that two debris flows were found in sediments deposited before 1945 and one after 1963. There was no report of flood or earthquake before 1945. Meanwhile, a heavy rainfall disaster (Nagasaki daisuigai) occurred at 1982 (Nagasaki city, 1989), which caused landslides and debris flow around the reservoir (Nishiyama and Chigira, 2002). The sand layer at the depth above 322 cm was, therefore, related to this disaster in 1982.

A peak of macroscopic charcoal was observed at 448 – 450 cm and those of microscopic charcoal were observed at 355 – 358 cm, 444 – 446 cm and 491 – 493 cm. The peak of macroscopic charcoal and that of microscopic charcoal at 444 – 446 cm were situated directly below the peaks of $^{239+240}$Pu and ^{137}Cs concentrations. It was reported that stratigraphic level with abundant macroscopic charcoal is inferred to be a proxy of local fire event, while that of microscopic charcoal reflected local and regional one (Whitlock and Larsen, 2001). The deposits of the charcoals in the sediments (444 – 446 cm and 448 – 450cm) might be a result of 'black rain' caused by fire after the detonation of the atomic bomb, which poured around the Nishiyama area after 20 minutes from the detonation (The Committee for the Compilation of Materials on Damage Caused by the Atomic Bombs in Hiroshima and Nagasaki, 1981).

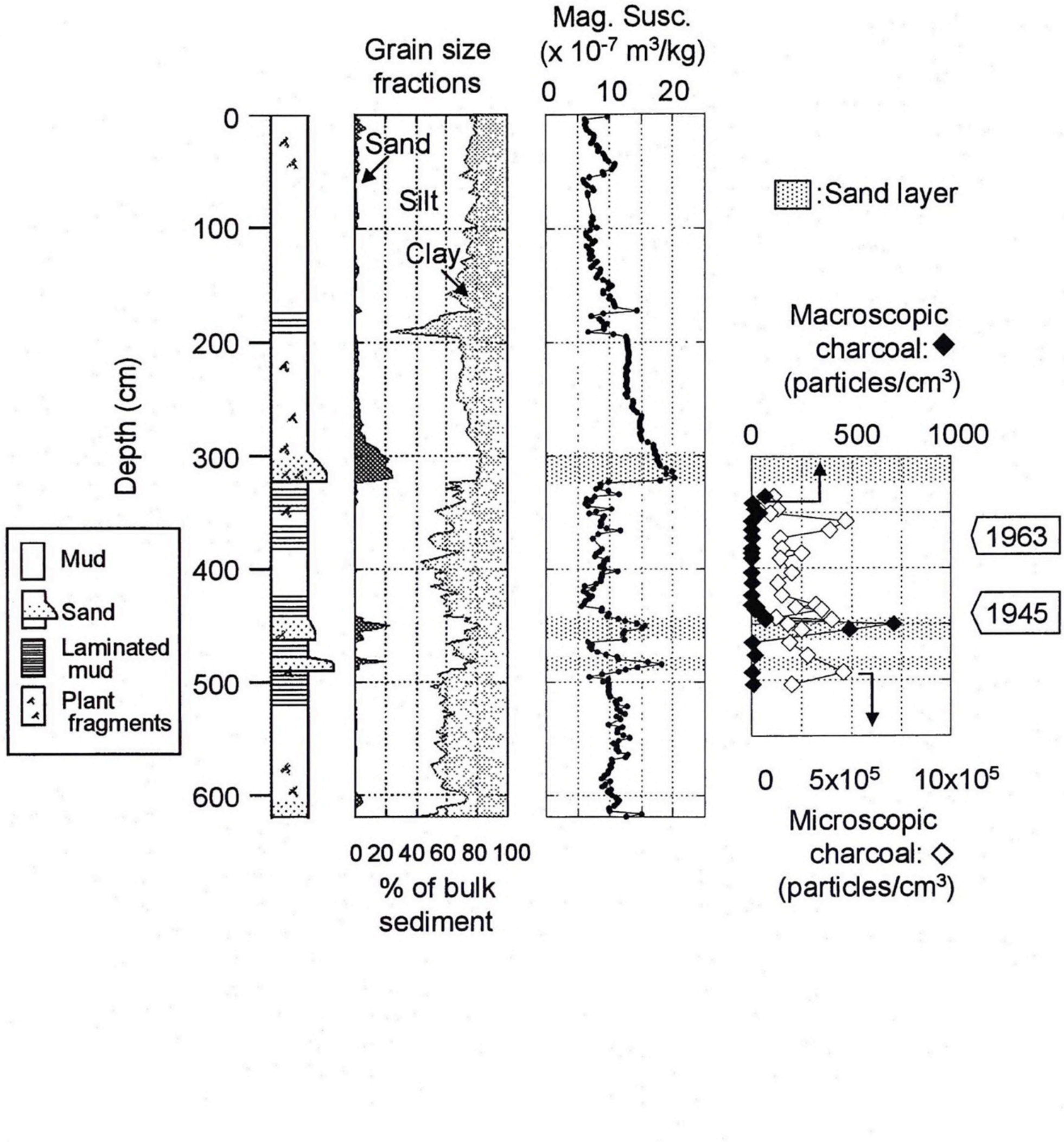

Fig. 3-3 Detailed sedimentology (lithofacies, grain size and magnetic susceptibility) and depth profiles of charcoal concentrations with chronological markers in the C core.

4. Spatial distribution of plutonium released from Nagasaki atomic bomb in surface soil

4.1 The spatial distribution within a 10 km radius of the hypocenter in Nagasaki city

In Chapter 3, the results of $^{240}Pu/^{239}Pu$ ratios made it clear that the soils flow into the reservoir and continue to supply the plutonium from the atomic bomb even now. However there is hardly any report about surface plutonium distribution, from the view point of $^{240}Pu/^{239}Pu$ ratios, in soils collected around the hypocenter containing the Nishiyama area. Surface soil samples were collected within a 10 km radius of the hypocenter and the $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ concentration were measured in order to determine spatial distribution of plutonium from the atomic bomb. The distribution of ^{137}Cs was also obtained to compare with that of plutonium.

The obtained values of the $^{240}Pu/^{239}Pu$ ratio and the $^{239+240}Pu$ and ^{137}Cs concentrations in soil (1) and (2), together with the ^{137}Cs concentrations measured by Noritake et al. (2002), are given in Table 4-1. The isotopic ratios were found to be between 0.032 and 0.22, while $^{239+240}Pu$ and ^{137}Cs concentrations were 0.025 – 23 Bq/kg and less than 1.9 to 84 Bq/kg, respectively.

Figure 4-1 shows the $^{240}Pu/^{239}Pu$ ratios at the sampling points. They are classified, according to isotopic ratios, into three groups. The isotopic ratios in the soils of the eastern area of the hypocenter (E1 – E12 except E8 and E11) were significantly lower than global fallout value (0.176 ± 0.014) (Krey et al., 1976). The mean value of the isotopic ratios in the soils around the Nishiyama reservoir (E1 – E7 and E9) was 0.038 ± 0.007. This value is close to 0.042 ± 0.014, 0.020 – 0.034 and 0.037 ± 0.002 reported by Komura et al. (1984), Yamamoto et al. (1985) and Muramatsu et al. (2003), respectively, for soils in the Nishiyama area. It is indicated, therefore, that the plutonium from the atomic bomb was dominant component deposited in the soils. The isotopic ratios in the soils were almost constant up to distances of 6 km (0.03 – 0.04), and then increased with distance. Even at 8 km from the hypocenter, the $^{240}Pu/^{239}Pu$ ratio was lower than the global fallout value. This showed a possibility that the plutonium from the atomic bomb was dispersed still further away. Obviously, it is necessary to analyze samples further eastern area to identify more precisely

Table 4-1 Isotopic ratio of $^{240}Pu/^{239}Pu$ and concentrations of $^{239+240}Pu$ and ^{137}Cs in soil (1) and (2) collected within a 10 km radius of the hypocenter in Nagasaki city

(Error :2SD)

Location Site No.	Distance from hypocenter (m)	$^{240}Pu/^{239}Pu$ ratio	$^{239+240}Pu$ (Bq/kg)	^{137}Cs (Bq/kg)	Sample No.*
E1	1975	0.049 ± 0.005	1.3 ± 0.2	29 ± 1*	1510
E2	2340	0.032 ± 0.003	22 ± 4	73 ± 2	
E3	2760	0.032 ± 0.001	16 ± 3	32 ± 2	
E4	3025	0.039 ± 0.009	2.0 ± 0.4	13 ± 1	
E5	3300	0.033 ± 0.001	23 ± 4	50 ± 2	
E6	3625	0.033 ± 0.001	4.2 ± 0.7	15 ± 1	
E7	4700	0.046 ± 0.002	10 ± 2	84 ± 2*	1405
E8	4850	0.17 ± 0.02	1.0 ± 0.02	43 ± 2*	1409
E9	5650	0.042 ± 0.004	1.9 ± 0.3	12 ± 1*	1404b
E10	6225	0.11 ± 0.02	0.50 ± 0.09	16 ± 1*	1407
E11	7550	0.17 ± 0.05	0.21 ± 0.04	10 ± 1*	1402
E12	7925	0.13 ± 0.01	1.7 ± 0.3	78 ± 2*	1401
E13	8175	0.17 ± 0.02	1.5 ± 0.2	60 ± 2*	1403
N1	900	0.18 ± 0.08	0.025 ± 0.006	< 1.9 **	1603
N2	2375	0.14 ± 0.02	0.46 ± 0.07	18 ± 1*	1410
W1	3275	0.17 ± 0.05	0.13 ± 0.03	7 ± 1*	1604
W2	4300	0.17 ± 0.01	0.60 ± 0.09	23 ± 1*	1514
W3	6100	0.22 ± 0.07	0.12 ± 0.03	4 ± 1*	1606
S1	850	0.17 ± 0.06	0.12 ± 0.03	6 ± 1*	1601
S2	3675	0.17 ± 0.01	0.9 ± 0.1	28 ± 1*	1501

* Noritake et al., 2002. **Yamazaki, 2005.

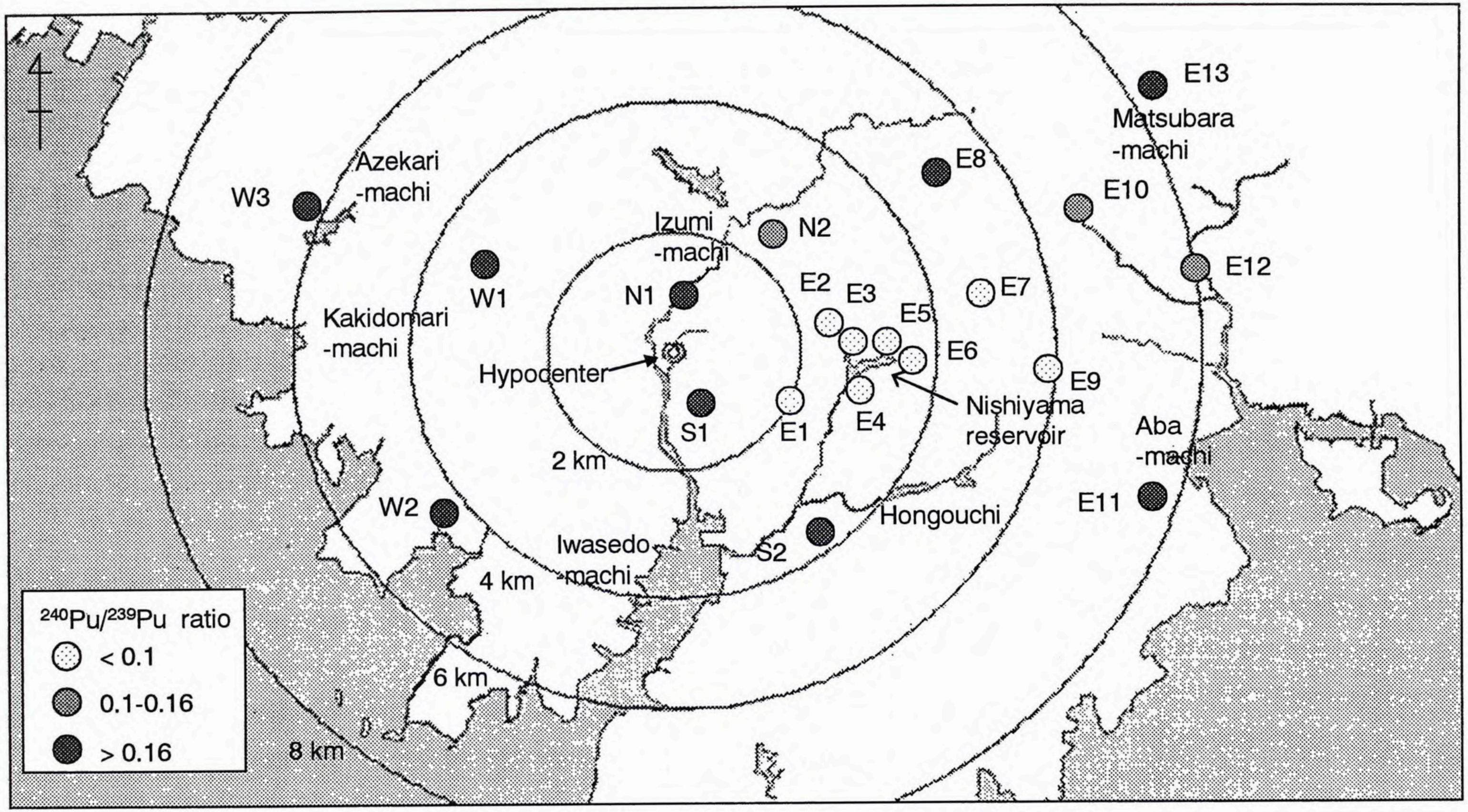

Fig. 4-1 Distribution of $^{240}Pu/^{239}Pu$ ratio in soils collected within a 10 km radius of the hypocenter in Nagasaki city.

the extent of the area which plutonium released from the atomic bomb was deposited. On the other hand, the isotopic ratio which was found in soil samples taken from around the hypocenter and in the northern, western and southern areas of the hypocenter ranged from 0.14 to 0.22 close to the corresponding global fallout value and the value (0.17 – 0.26) in depositions collected at Nagasaki city in 2000 (Hirose et al., 2003). From these results, it can be concluded that the plutonium from the atomic bomb was dispersed over the eastern area and then deposited in the soils there, especially in the Nishiyama area.

Figure 4-2 shows correlation between $^{239+240}Pu$ and ^{137}Cs concentrations in the samples collected at the eastern area. In addition to the data of this study, the concentrations and the average values for 18 soil samples taken from uncultivated lands at the depth of 0 – 5 cm all over Japan (Japan Chemical Analysis Center, 2002) are also plotted together with their average value. The mean activity ratio of $^{239+240}Pu/^{137}Cs$ in the soils was 0.028 ± 0.08 equivalent to a typical global fallout level of 0.031 (decay corrected to 2002) reported by Hardy et al. (1973). The data were divided into two groups. Seven samples, E2 – E7 and E9, form 'Group A' and the rest of the samples form 'Group B'. Statistical analysis was carried out at 95% significance level to investigate the differences between variables of the study groups. Statistical significance was concluded when p-value was < 0.05. With Welch's t-test ($p=0.0056$), significant difference was found in the $^{239+240}Pu/^{137}Cs$ activity ratios of the study groups. The $^{240}Pu/^{239}Pu$ ratios in the samples of 'Group A' were about one-fifth as low as that in the global fallout. The $^{239+240}Pu/^{137}Cs$ activity ratio in the samples also indicates that the origin of plutonium is the Nagasaki atomic bomb. On the other hand, the $^{239+240}Pu/^{137}Cs$ activity ratios (0.022 – 0.045) in other samples of 'Group B' were comparable to those in the soils collected in the other areas of Japan reported by Japan Chemical Analysis Center (2002). However, samples E1, E10 and E12 had lower $^{240}Pu/^{239}Pu$ ratios. If the $^{239+240}Pu/^{137}Cs$ activity ratio alone is used to identify the origin of plutonium, then it is concluded that the radionuclides in the samples of 'Group B' are originated from global fallout. However, the $^{240}Pu/^{239}Pu$ ratio in these samples was lower than that of global fallout. It clearly indicates that

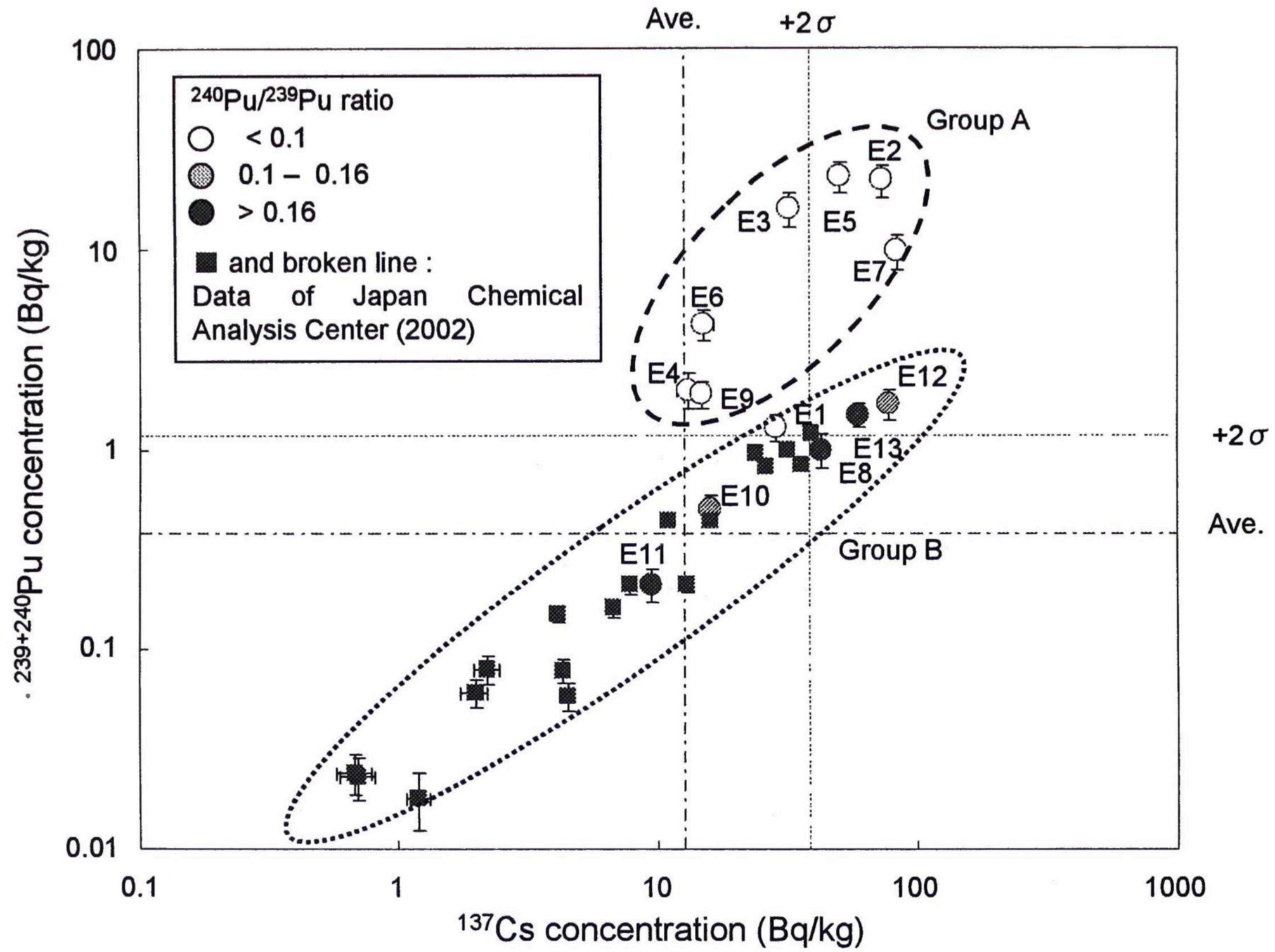

Fig. 4-2 Correlation of ^{137}Cs concentration to $^{239+240}$Pu concentration in soils collected at the eastern area in Nagasaki city. (Error: 2SD).

the soils contain plutonium from the atomic bomb. Therefore, the $^{240}Pu/^{239}Pu$ ratio is more effective in the identification of plutonium origin than the $^{239+240}Pu/^{137}Cs$ activity ratio or the $^{239+240}Pu$ and ^{137}Cs concentrations itself, especially for the case of low plutonium concentration.

4.2 The spatial distribution in eastern area of Nagasaki city

In order to determine the entire distribution of plutonium from the atomic bomb, the study area was expanded toward the eastern area of Nagasaki city.

The obtained data of the $^{240}Pu/^{239}Pu$ ratios, the $^{239+240}Pu$ and ^{137}Cs concentrations in the soil (3) are given in Table 4-2, together with the data from the previous section (E2, E12 and W3 is renamed N1, N2 and N7) and the data (N*, K* and O*) reported by Isogai (2005). Figure 4-3 shows the $^{240}Pu/^{239}Pu$ ratios at the sampling points. The isotopic ratios were ranged between 0.033 and 0.22. Isotopic ratios of 0.033 – 0.15 were observed in the samples N1, N2, N5, N*, N6, K4, P5, P6 and K*. These values were relatively lower than the global fallout value. The area where the low ratio was observed covers wide eastern area from the hypocenter including the Nishiyama area, the Shimabara Peninsula and a part of northern area in Kumamoto prefecture. In contrast to these findings, the $^{240}Pu/^{239}Pu$ ratios in the samples from other directions were 0.15 – 0.22, which are comparable to the global fallout value. It indicates that the lower $^{240}Pu/^{239}Pu$ ratios in the samples of N1, N2, N5, N*, N6, K4, P5, P6 and K* were influenced by the plutonium from the atomic bomb. The plutonium from the atomic bomb was found the area between 2 km and about 100 km east from the hypocenter and about 30 km north and south wide. The area was larger than that reported in the previous studies (Yamamoto et al., 1985; Kudo et al., 1991).

It was reported that a cumulonimbus cloud formed immediately after the detonation was drifted toward a northeasterly direction by wind with a velocity of about 3 m/s (The Committee for the Compilation of Materials on Damage Caused by the Atomic Bombs in Hiroshima and Nagasaki, 1981) and passed over the Unzen Volcanic Mountain

Table 4-2 Isotopic ratio of $^{240}Pu/^{239}Pu$ and concentrations of $^{239+240}Pu$ and ^{137}Cs in soil (3) collected in north part of Kyushu area

(Error :2SD)

Pref.	Sample ID	$^{240}Pu/^{239}Pu$ ratio	$^{239+240}Pu$ (Bq/kg)	^{137}Cs (Bq/kg)
Nagasaki	N7(W3)**	0.22 ± 0.07	0.12 ± 0.03	4 ± 1
	N1(E5)**	0.033 ± 0.001	23 ± 4	50 ± 2
	N2(E12)**	0.13 ± 0.01	1.7 ± 0.3	77 ± 2
	N3	0.2 ± 0.2	0.013 ± 0.008	30 ± 7
	N4	0.18 ± 0.1	0.30 ± 0.05	14 ± 2
	N5	0.15 ± 0.02	1.4 ± 0.2	34 ± 4
	N6	0.111 ± 0.004	2.0 ± 0.3	12 ± 4
	P1	0.20 ± 0.05	0.19 ± 0.04	-
	P2	0.19 ± 0.04	0.45 ± 0.09	-
	P3	0.21 ± 0.06	0.22 ± 0.05	-
	P4	0.18 ± 0.04	0.52 ± 0.09	-
	N*	0.13 ± 0.02	1.3 ± 0.1	-
Kumamoto	K1	0.17 ± 0.06	0.06 ± 0.01	0.8 ± 1.4
	K2	0.17 ± 0.01	0.45 ± 0.07	21 ± 5
	K3	0.19 ± 0.01	0.37 ± 0.05	23 ± 3
	K4	0.105 ± 0.009	1.7 ± 0.3	48 ± 4
	K5	0.22 ± 0.05	0.06 ± 0.01	3 ± 3
	K6	0.19 ± 0.03	0.1 ± 0.2	7 ± 2
	P5	0.12 ± 0.06	0.08 ± 0.02	-
	P6	0.15 ± 0.22	0.03 ± 0.03	-
	P7	0.20 ± 0.02	0.23 ± 0.03	-
	P8	0.17 ± 0.03	0.19 ± 0.03	-
	P9	0.17 ± 0.06	0.05 ± 0.01	-
	P10	0.18 ± 0.02	1.4 ± 0.2	-
	P11	0.18 ± 0.03	0.40 ± 0.07	-
	P12	0.17 ± 0.04	1.0 ± 0.2	-
	P13	0.18 ± 0.15	0.05 ± 0.03	-
	P14	0.20 ± 0.07	0.37 ± 0.08	-
	K*	0.12 ± 0.02	4.1 ± 0.6	-
Ohita	O1	0.18 ± 0.02	0.7 ± 0.1	19 ± 3
	O2	0.19 ± 0.03	0.43 ± 0.07	10 ± 5
	O3	0.18 ± 0.02	0.8 ± 0.1	24 ± 5
	O4	0.18 ± 0.09	0.09 ± 0.02	4 ± 4
	O5	0.18 ± 0.01	1.5 ± 0.2	32 ± 23
	O6	0.19 ± 0.02	0.8 ± 0.1	37 ± 17
	P15	0.18 ± 0.02	0.7 ± 0.1	-
	O*	0.17 ± 0.06	2.7 ± 0.5	-
Saga	S1	0.17 ± 0.05	0.029 ± 0.007	3 ± 2
	S2	0.18 ± 0.03	0.21 ± 0.03	10 ± 4
	S3	0.17 ± 0.02	0.40 ± 0.06	18 ± 2
	P16	0.19 ± 0.02	0.18 ± 0.03	-
	P17	0.18 ± 0.06	0.21 ± 0.04	-
Fukuoka	F1	0.15 ± 0.04	0.14 ± 0.03	7 ± 2
	F2	0.18 ± 0.04	0.50 ± 0.07	22 ± 10
	P18	0.19 ± 0.03	0.29 ± 0.05	-
	P19	0.16 ± 0.02	0.15 ± 0.02	-

-: no data.

* Isogai, 2005.

** The samples of W3, E5 and E12 in soil (1) and (2) were renamed N7, N1 and N2.

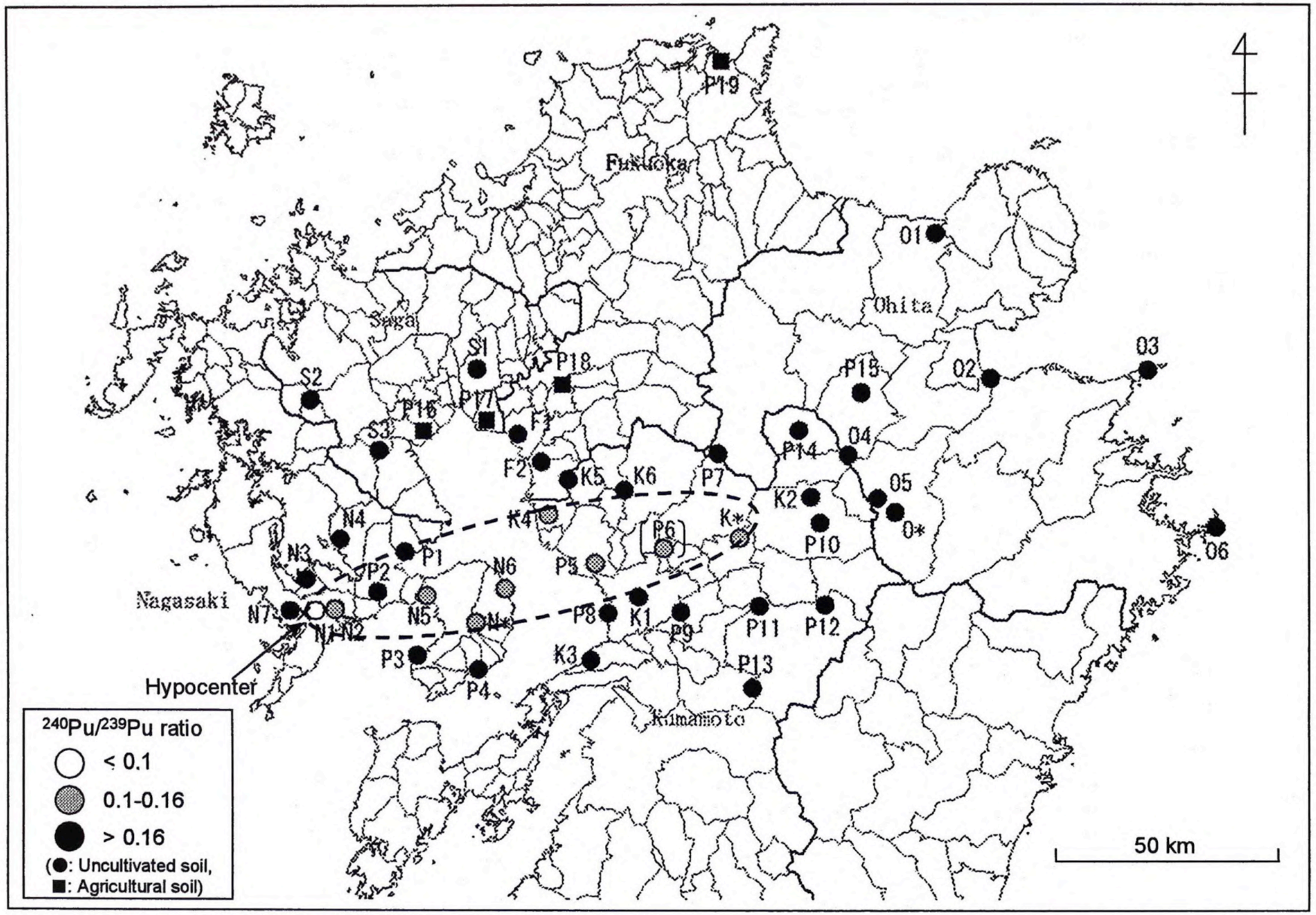

Fig.4-3 Distribution of $^{240}Pu/^{239}Pu$ ratio in soils collected in the eastern area of Nagasaki city.

Data in samples with superscript of * was reported by Isogai (2005)

** Data of P6 was a information value, because the uncertainty was large.

Meteorological Station, which is about 45 km east of the hypocenter (Ishida, 1953). During the initial period, 20 minutes after the detonation, the 'black rain' fell onto the Nishiyama area (The Committee for the Compilation of Materials on Damage Caused by the Atomic Bombs in Hiroshima and Nagasaki, 1981). The area where the plutonium deposited in Nagasaki prefecture was consistent with area which the cloud of the atomic bomb passed over and the area in Kumamoto prefecture was downwind from the area in Nagasaki prefecture. The plutonium from the atomic bomb in the cloud might be carried to Kumamoto prefecture through Nagasaki prefecture by the wind.

In Nagasaki prefecture, areas where the soil has low $^{240}Pu/^{239}Pu$ ratio were similar to those where high intensity of γ-ray in soils were detected a few months after the detonation (Nakane, 2000). The plutonium from the atomic bomb was not found at P2 which was located at about 20 km east from the hypocenter, although the plutonium was detected at N1 and, N5, N* and N6 which was further eastern area than P2. It was reported that the cloud which caused rain to fall on the Nishiyama area reached Unzen which locates between N5 and N6 and a slight rain fall began about three hours after the detonation (Ishida, 1953). From these results, it is possible to say that the precipitation contributed the local fallout.

Moreover the $^{240}Pu/^{239}Pu$ ratio in N6 was lower than that in N5 and, which suggested that amount of the plutonium from the atomic bomb deposited in N6 was more than that in N5. The intensity of γ-ray in area around N6 was also higher than that around N5. One of the probable reasons was geographical features; the area around N6 is flatter than that around N5.

The $^{239+240}Pu$ and ^{137}Cs concentrations ranged 0.013 – 23 Bq/kg and 0.8 – 77 Bq/kg (Table 4-2). However, data in some samples cannot be directly compared with those of the other samples for the following reason: the soil samples (N2 and N7) were collected at depths ranging from several centimeters to 20 centimeters below the surface. All other soil samples were collected at depths from 0 – 5 cm or 2 – 5 cm. Mahara et al. reported that soil at the depth of 0 – 10, 10 – 20 and 20 – 30 cm retained 90%, 5% and 2% of the total $^{239+240}Pu$ content and 96%, 2% and 1% of the total ^{137}Cs content in soils at the Nishiyama area,

respectively (Mahara and Miyahara, 1984; Mahara and Kudo, 1995). Moreover the samples (P16 – P19) were collected in agricultural fields. The depth profile of ^{137}Cs in cultivated soils is constant within the plough layer which is mixed the soil and the associated ^{137}Cs by the tillage practices (He and Walling, 2000; Fukutani and Takahashi, 2003). For these reasons, the concentrations in the soils (N2, N7 and P16 – P19) may be underestimated when compared with those of the other soils. This fact is considered in the following discussion.

Figure 4-4 shows the $^{239+240}Pu$ and ^{137}Cs concentrations in soils with distance from the hypocenter. Data in soils of E2, E8 and E10 were also shown to obtain the detailed distribution. High levels of $^{239+240}Pu$ were observed in area which located about 2 – 3 km east from the hypocenter, while those of ^{137}Cs were in area of about 2 – 8 km east from the hypocenter. It indicated that plutonium and ^{137}Cs from the atomic bomb were deposited in the areas. However the results of $^{240}Pu/^{239}Pu$ ratio showed the plutonium from the atomic bomb was carried to about 100 km east from the hypocenter. It is, therefore, revealed that the $^{240}Pu/^{239}Pu$ ratio is powerful tool to determine the spatial distribution of plutonium from the atomic bomb, even if plutonium concentration is low.

The spatial distribution of plutonium was different from that of ^{137}Cs, a fission product of plutonium. The results of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ concentration indicated that the plutonium was deposited in local area, especially in the Nishiyama area. Meanwhile, the deposition of ^{137}Cs was broader than plutonium. Kudo et al. (1991) pointed out the same distributions from the results of $^{239+240}Pu$ and ^{137}Cs concentrations. It is probably due to difference in the production process during their transportation. Plutonium is a refractory element without any gaseous precursors and existed as solid particles, whereas ^{137}Cs is a fission product with gaseous precursors: $^{137}Te \rightarrow {}^{137}I \rightarrow {}^{137}Xe \rightarrow {}^{137}Cs$. The spatial distributions provide opportunities to understand environmental behavior of plutonium and ^{137}Cs released from the atomic bomb.

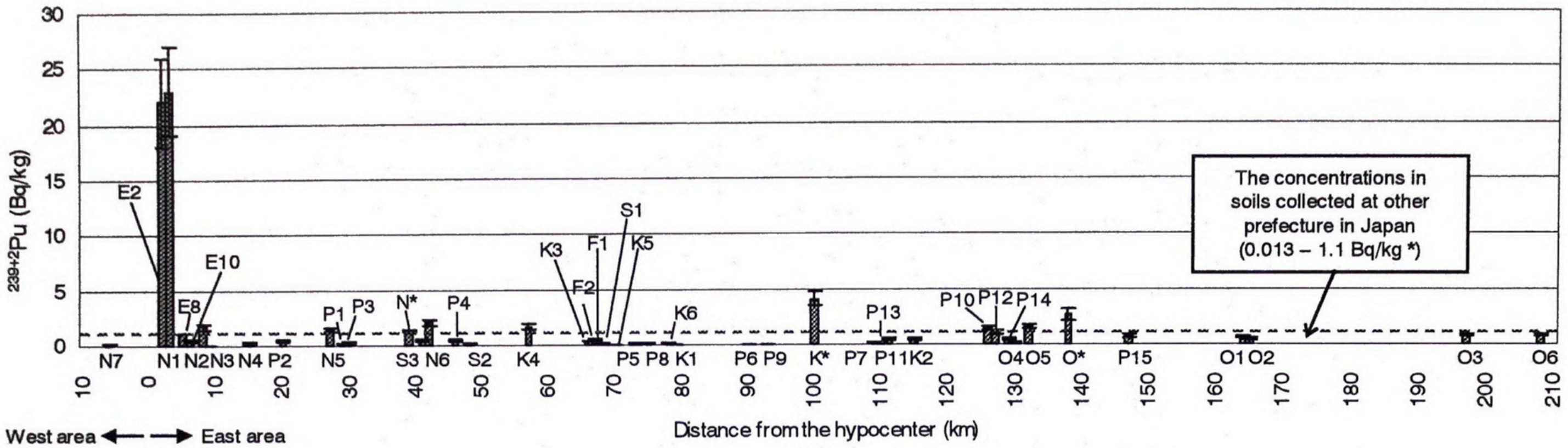

Fig. 4-4 The $^{239+240}Pu$ and ^{137}Cs concentrations in soils with distance from the hypocenter.

* Japan Chemical Analysis Center, 2002.

** Except P16-P19.

5. Conclusions

Temporal and spatial distribution of plutonium with ^{137}Cs released from Nagasaki atomic bomb was investigated from $^{240}Pu/^{239}Pu$ ratio with $^{239+240}Pu$ and ^{137}Cs concentrations in sediment cores collected at Nishiyama reservoir and those in surface soil collected around the hypocenter and at eastern area of Nagasaki city. Measurement of $^{240}Pu/^{239}Pu$ ratio played the decisive role in the identification of plutonium origins and enabled us to discriminate plutonium due to the detonation of Nagasaki atomic bomb from that of atmospheric nuclear tests.

Firstly, depositional records of the plutonium and ^{137}Cs from the atomic bomb have been obtained from depth profiles of $^{240}Pu/^{239}Pu$ ratio together with $^{239+240}Pu$ and ^{137}Cs concentrations in sediment cores collected at Nishiyama reservoir. From the results of the C core, the release of the plutonium by the detonation caused a sharp peak of the $^{239+240}Pu$ deposits in a specific sediment. Then the deposits have been decreased with the passage of time. However a peak of $^{240}Pu/^{239}Pu$ ratio found in a sediment above that shown $^{239+240}Pu$ peak indicated deposit of plutonium from atmospheric nuclear tests. Even at present, the plutonium from the atomic bomb is continuously being deposited dominantly. The $^{240}Pu/^{239}Pu$ ratios in the sediment from surface to 301 cm in C core are comparable to those in soils around the reservoir. The soils have been a source of the plutonium supply for the sediments a long period of time. On the other hand, deposition of ^{137}Cs from the atomic bomb also formed a sharp peak of ^{137}Cs concentration. The depth profile of ^{137}Cs concentration was consistent to those of $^{230}Pu/^{230}Pu$ ratio and $^{239+240}Pu$ concentration, which indicated that the results of ^{137}Cs was complementary to those of plutonium.

From the depth profiles of plutonium and ^{137}Cs found in the C core, a chronological marker was fixed for 1945, that points the beginning of sedimentation of plutonium and ^{137}Cs from the atomic bomb, and another marker for 1963, maximum deposition from the nuclear tests. The C core had sediments which had been deposited for about 100 years and probably records some other events occurred in this term. In this study, trace of two other events were observed; Nagasaki daisuigai (a heavy rain disaster) and a fire occurred by the detonation of

the atomic bomb. If depth profiles of other substances such as heavy metal, dioxin and diatom in the C core are determined, it might provide information of chronological trends of other contaminations.

Secondly, data of $^{240}Pu/^{239}Pu$ ratio and $^{239+240}Pu$ and ^{137}Cs concentrations in surface soils collected in wide area covering northern part of Kyusyu area including the hypocenter and Nishiyama area were obtained to elucidate the distribution of the plutonium released from the atomic bomb. Lower $^{240}Pu/^{239}Pu$ ratio than global fallout value was found from the eastern area of the hypocenter in Nagasaki city to a part of the northern area in Kumamoto prefecture. The $^{239+240}Pu$ and ^{137}Cs concentrations in the area except the surrounding areas of Nishiyama were close to global fallout level, that is, background level in Japan. The results of the $^{240}Pu/^{239}Pu$ ratios showed that the plutonium from the atomic bomb was carried from the hypocenter toward the eastern area and also reached a part of the northern area in Kumamoto prefecture. The atomic bomb was observed between 2 km and about 100 km east from the hypocenter and about 30 km north and south wide. The area was larger than the contamination area reported in the previous studies.

The obtained temporal and spatial distribution of plutonium from the atomic bomb could contribute to the understanding of the behavior from the release to the sedimentation. In addition, the data coupled with more detailed weather record and information of the chemical/physical mechanism in the environment will enable us to assess the detailed behavior.

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Publication lists

Papers included in the thesis

1. "Geological distribution of plutonium isotopes of Nagasaki atomic bomb spread over Nagasaki and Kumamoto area"

   Saito-Kokubu, Y., Yasuda, K., Magara, M., Miyamoto, Y., Sakurai, S., Usuda, S., Yamazaki, H., Yoshikawa, S.

   Proceedings of the seventh workshop on Environmental Radioactivity, p.36-40 (2006).

2. "Plutonium isotopes derived from Nagasaki atomic bomb in the sediment of Nishiyama reservoir at Nagasaki, Japan"

   Saito-Kokubu, Y., Esaka, F., Yasuda, K., Magara, M., Miyamoto, Y., Sakurai, S., Usuda, S., Yamazaki, H., Yoshikawa, S., Nagaoka, S.

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3. "Distribution of plutonium isotopes and ^{137}Cs found in the surface soils of Nagasaki, Japan"

   Saito-Kokubu, Y., Yasuda, K., Magara, M., Miyamoto, Y., Sakurai, S., Usuda, S., Yamazaki, H., Mitamura, M., Yoshikawa, S.

   J. Geosci., Osaka City Univ., **50**, 7-13 (2007).

4. "Geographical distribution of plutonium derived from the atomic bomb in the eastern area of Nagasaki"

   Saito-Kokubu, Y., Yasuda, K., Magara, M., Miyamoto, Y., Sakurai, S., Usuda, S., Yamazaki, H., Yoshikawa, S.

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5. "Depositional records of plutonium and ^{137}Cs released from Nagasaki atomic bomb in sediment of Nishiyama reservoir at Nagasaki"

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Other papers

1. "Investigation of environmental migration and behavior by trace of Nagasaki atomic bomb recorded in sediments"
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